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Vertical nutrient and trace element migration in cambisols after application of residues from anaerobic digestion of manure

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Abstract

At an alpine grassland cambisol, one site had been intensely fertilized with urban sewage sludge for 10 years of 7,5 tons/ha annually, whereas an adjacent site had been left untreated. A model column experiment was set up to investigate changes of permeabilities and trace element retentions at 0-20 cm and 20-60cm layers thereof. The particular goal was to monitor losses of nutrients to deeper soil layers or aquifers from biogas residues deposited on site. Residual slurry after biogas production or equivalent amount of water were added on top of the model columns, followed by gradual elution with de-ionized water at amounts of expected rainfall at the sampling site (1000 mm). Long-term sludge treatment changed organic carbon, soil nutrients as well as Ba, Cu, Pb, and Zn significantly, resulting in different vertical migration and elution of applied substances. Differences between the mobilities of P, Fe, S, Cu, B, as well as nitrate and ammonia losses were much more affected by the long-term sludge pretreatment than by the characteristics of the biogas residue or just de-ionized water applied on top of the columns. Largest differences between biogas residue addition and water only were noticed for zinc and ammonia. The addition of iron salts in order to increase sulfide precipitation, had measurable but low effects.

Keywords: sewage sludge deposition, vertical mobilities in soil, soil columns, phosphate, sulfate, nitrate, ammonium, Fe-treatment of biogas residues

1. Introduction

Soil has been defined as a zone of microbial life, a substratum for green plants, and a habitat for animals. Soil classification schemes have been developed due to the succession of soil layers with increasing depth, which have been characterized by chemical, physical and microbial properties. The competence of geology starts at the depth where soil life ends, which is clear in case of underlying rocks, but it is an important definition for underlying porous sediments.

1.1 Soil fertility

In the past, major subjects of agricultural research aimed at soil fertility increases by optimization of the N- and P- supply to arable crops, and at the prevention of substantial losses of nitrate, ammonium, and phosphate to the groundwater and to surface waters. In recent years, degradation of the organic carbon pool due to continuous mineral fertilization and growing crops of low root biomass, has attracted increased attention worldwide, such as in Iceland and the East of Austria (Van Leeuwen et al. 2015), Italy (Macci et al. 2013), Africa (Batjes 2014), India (Debasih-Saha 2014), Brazil (Fialho and Zinn 2014) and elsewhere.

1.2 Effects upon groundwater composition

The formation of groundwater below soils is largely governed by filtering and release of solutes in the soil column. Lysimeter and column studies are valuable tools to investigate

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these processes. Below the zone of soil live, nutrients are not needed, and thus remain. In addition, adsorption or degradation of contaminants in aquifers is low. In general, recycling of organic wastes by application to soils has caused concerns about impacts on groundwater quality. In order to achieve nutrient cycling within agriculture, losses to groundwater, potable water and surface waters should be avoided (Sager 2013).

1.3 Soil organic carbon

Soil organic carbon is the largest terrestrial active sink for atmospheric carbon, and serves as an indicator for water retention, physical stability, and retention of nutrients and essential micro-elements. Soil organic and adequate humidity are pre-conditions of microbial life, which governs the major reactions in the N and P-cycles. Increase in organic carbon stocks can be achieved from changes in tillage like covering bare soil or return of crop residues, use of composts and manures as fertilizers, and erosion control. After 30 years of field trials at Seehausen (Germany), organic fertilization significantly enhanced soil biological activities, like soil enzymes and abundance of soil organisms, increase in macropores, and decrease of density (Hülsbergen 2011).

Carbon can be added via compost, manure, sewage sludge, and urban biowaste. Whereas in some regions, consequent mineral fertilization of arable land and the end of keeping domestic animals, has gradually depleted organic carbon and thus soil life, the number of domestic animals kept in regions of permanent grassland has steadily increased, and thus the amount of manure produced there. The potential of compost production in these regions, however, is low because the pasture plants are largely fed to the animals. Manure can be used as an organic fertilizer without further treatment, but long-range transport is not economical.

1.4 Sewage sludge addition to soils

At first, sewage sludge was regarded as a source of nutrients applied to the soil. Later it was recognized that appreciable metal loads contributed to contamination, and simultaneous addition of organic matter with plenty of microbial life changed soil characteristics and groundwater formation processes. Beneath changes in redox potential and pH, this leads to changes in mobilities in the respective soil column and percolate composition, as well as changes in mobilities obtained by selective leaching methods in the batch mode. Contrary to mobile fractions obtained in batch mode, microbial life is still going on, and chemical equilibrium can be achieved only in each differential layer, but not across the entire column. This is more close to conditions encountered in nature, but much more work also.

When stabilized sewage sludge was added to columns of coarse sand, medium sand, or coal fly ash, and washout was performed with water equivalent to heavy rainfall, breakthrough of nitrate as a conservative tracer occurred within 1 day. Ammonium retardation was 2 times, retardation of organic nitrogen compounds was 1,5 times more than retardation of nitrate. Whereas sewage sludge phosphate was almost completely recovered in the eluates, release of metals was largely below 1% of added amounts within 30 days, and stabilized at a constant low but measurable level (Łuckiewicz 2006). The same column devices were also used to monitor the transport of bacteria (Clostridium perfringens and fecal coliforms). Straining,

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adsorption and inactivation are the main processes controlling bacterial transport in porous media. From the sandy columns, the breakthough of bacteria occurred later than sludge-born nitrate, whereas below fly-ash, no breakthrough occurred within 30 days. Bacterial movement in the sandy columns was similar to organic N and ammoium, while there was no such correlation in the fly ash, probably because of its alkaline pH, sorption capabilities, and metal contents (Łuckiewicz, Quant 2007).

In case of sewage sludges, deposition to permanent grassland has been regarded as a sound waste management practice, because the harvest from these sites is hardly suitable for direct human nutrition. Biogas produced from manure provides additional income, either because of lowering the need for fossil fuels, selling to natural gas suppliers, or direct transformation into electricity. For more than 10 years, more than 300 facilities have been working in Austria to produce biogas.

1.5 Manure addition to soils

Downward migration of substances added as manure resp. biogas residues cannot be concluded from experiments with solutes, because of aggregates and colloidal forms. Size exclusion chromatography of membrane filtrated liquid of a pig slurry of pH=8 showed a bimolecular distribution of organics due to molecular weight, with peaks at the low molecular fraction, and another at high molecular mass humic substances. In the fraction <1000 D, many low molecular fatty acids could be identified like acetic, propionic, butyric, and valeric acids, but also oxalate, but no lactate. One third of the Cu and Zn was found in the mass range > 50000 D. Whereas Cd and Zn complexes dissociated rapidly in presence of a solid exchanger, Cu complexes were stable (Del Castilho et al. 1993).

Cattle slurry contains nitrate, ammonium and partly phosphate in solution, whereas organic matter is present in dissolved, colloidal and solid states. When cattle slurry was spread to bare soil, infiltration velocity, retention at solid soil particles wass thus different. After addition of cattle slurry to soil columns made of 25 cm of A-horizon and 25 cm of B-horizon, about 10 slurry volumes of water were needed, till there was no appreciable washout from the slurry to the soil under saturated moisture condtions. Release of additional ammonium from mineralization of organic nitrogen was observed. Clay minerals present in the soil slowly adsorbed potassium from the slurry (Lopez Periago et al. 2000). Whereas from soil columns treated with water, 65-98% of P in the eluates was dissolved, colloidal P in the effluents from swine-manured soils was 26,7 times higher, and dissolved P smoothly increased during the breakthrough experiment. Good linear corrrelation between colloidal P and colloidal Fe in the eluates indicated that Fe hydroxides transport colloidal P (Zang et al. 2011).

1.6 Metal contaminations

Most respective papers are focused on C and N only, but do not care about the input of accessory elements contained in manures and composts, which is significantly higher than from mineral fertilizers in most cases, particularly based on nutrient N or P contents, including even cadmium (Sager 2007, Sager 2011). In particular, urban biowaste occasionally

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contains contaminants at significant levels and has to be intensely controlled, which raises the costs of application (Sager 2012).

Application of sludge to an agricultural soil west of Paris (France) led to enrichment of heavy metals in the first 40 cm and also to an increase in organic matter (8%) and carbonates, whereas Fe remained constant in the soil profile, and Mn got depleted. The lower sandy horizon below the sludge –spread zone still contained higher levels of various metals and also more organic carbon. Though sewage spreading was stopped 55 years ago, and 30 years of cultivation and 25 years of meadow growing, groundwater formed at the site had 27-53% O_2-saturation and contained also enhanced metal levels, but below thresholds (Bourg 1995).

In sewage sludge, main fractions of Cd, Ni, and Zn were found in the moderately reducible fraction (NH_2OH- extract), which usually releases the Mn-oxides (Dudka, Chlopecka 1990).

When columns filled with 80 cm soil or 40 cm composted sewage sluge + 40 cm soil were eluated with dilute HNO_3: H_2SO_4 = 3:7, maximum Cu and Ni in the eluates appeared before the breakthrough of acid, at pH 5,3. This should imitate acid rain. In the combined profile, the metals Pb-Zn-Ni-Cu were transported from the compost layer to the soil horizon and retained there. Sequential leaching after the column experiment revealed that only exchangeable and weak-acid-mobile (termed carbonate) forms were transported through the profile, whereas others were left untouched (Kowalkowski, Buszewski 2009).

Ryegrass (lolium perenne L.) and white clover (trifolium pratense L.), grown at a lysimeter filled according to natural genetic horizons to a depth of 80cm, and 20 cm sewage on top, took 4-9 times higher metal loads than collected in the percolates below the columns; just for Cd, leaching was 5 times higher. Mean values of percolate waters from sludge treatment doubled within 3 years. Atmospheric fallout was negligible compared with sludge input (Dudka, Chlopecka 1990).

1.7 Biogas production

Manures and residues from biogas production usually contain more K, N, P, and S than soils, also more B and Mo, and some of them may be contaminated with Cu, Zn and Se. Methane formation in biogas facilities lowers the carbon content of the residues and thus the reproduction potential for humus, when applied as organic fertilizers to soils (Zimmer 2009).

In Germany, manure has contributed 20% to the overall biogas production, whereas 22% have come from the „green tons“, 24% from proper energy green plants, and 14% from food residues (Thelen-Jüngling 2011). Maximum amounts of 855 mg/kg for Cu, 1353 mg/kg for Zn and 2,47 mg/kg Se per dry mass were found in Austria in 2003 (see also Sager 2007). Together with the accumulation of persistent organic compounds, this will be probably the task of the next generation of researchers.

Prior to burning in a local electric power plant, or selling to natural gas suppliers, sulfur containing components (e.g. H_2S) in the biogas residues have to be kept at low levels, because the combustion product SO_2 is highly toxic and corrosive. Addition of divalent Fe-salts should keep all sulfur compounds in the biogas residue, finally precipitated as pyrite.

1.8 Choice of site and substrate of this work

This work shows differences in retention of solutes from manure applied to original and sewage-modified soil in column experiments. Contrary to previous column experiments (Sager 2002, Sager 2004), nutrient and trace elements were not applied as soluble salts on top of the columns, but as biogas residue, to simulate organic fertilization. This paper addresses long-term changes of an alpine grassland soil by sludge application with respect to chemical composition, changes of mobility, and retention of various items contained in manure residues after biogas production.

In some 100m distance from the sampling site, field lysimeters have been operated with local soils and various combinations of crops and fertilization regimes for more than 30 years. In particular, a carbonaceous pararendzina similar to the soil used in this paper, was treated in field lysimeters within 1981-1990, in order to investigate nitrification below various crops and fertilization regimes. This lysimeter soil had a pH of 7,5 and 4,7% humics content, as well as 21% sand, 68% silt and 14% clay. Aeration during the filling of theses lysimeters increased transformation of organically bound soil-N to yield increased nitrate washout within the first 2 years. Fertilization with manure decreased the eluate volumes, and increased the elution of ammonium, K, and Na. The annual P-output was smaller than 0,5kg P/ha except for the highest load of 480 kg N/ha, which significantly exceeds the current threshold of 170 kg N/ha (Bohner, Eder 2011).
The novelty of the column experiments in the lab with a similar soil, which is presented in this work, was to investigate the effect of the addition of $FeCl_2$ to fix the H_2S during biogas production from manures, to indicate mobility changes in the solid, and to extend the spectrum of investigated elements to others than C, N, P, K, and Na. A second item is to test the difference between biogas manure application at an untreated site, compared with a sludge- pretreated site of the similar soil.
The scope of this work has been to investigate possible changes of migration of substances applied as biogas residues, from long term sludge application, as well as the effect of added divalent iron salts to precipitate pyrite in the biogas residues, on P and trace element mobilities as well as nitrification reactions. Mobile fractions in soil column slices obtained after the experiment should monitor mobile phases and the downward migration of added P, Fe and others in the soil profile.

2. Material and methods

In order to address the problem of long-term changes of permanent grassland soils by long-time disposal of sewage sludge, two originally similar soils were sampled at 2 horizons from adjacent sites at the experimental station at Gumpenstein (Styria, Austria), which means 4 solid samples from 2 soils. They are of a non- carbonaceous cambisol type, consisting of an A-horizon of 20 cm and an AB-horizon 20-60 cm, which is quite common within the entire Ennstal alpine region, and makes 19% of the soils of the entire province of Styria. One permanent grassland-site had been fertilized with urban sewage sludge at 7,5t/ha for 10 years, and therefore will be called the „high P“ (P+ site), whereas the other had got no treatment and

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will be called „low P“ (P- site). The P- site had still enough organic carbon (classified as medium humics level) to ensure adequate microbial life. Soils from the same site like used in this work have been filled into field lysimeters already in 1991, and used for the investigation of nutrient losses below bare land and grassland under field conditions, after manure application (Eder 1997, Eder 1999).

For the column experiment presented in this work, plexiglass cylinders of 60 cm length and 12,6 cm diameter (125 cm²) were mounted in plastic Buechner funnels and closed with a nylon gauze at the bottom. The soil samples were air-dried, homogenized, and sieved <5 mm. The columns were filled with 22 cm soil from the AB-horizons, and then with 16,5 cm of the A-horizons, and wettened from top to bottom for 7 days to reach field capacity. After sacking, the column length before addition of the biogas residue was 35 cm.

The biogas manure had been produced from maize silage (25 t/d) and pig manure (20 m³/d) by anaerobic fermentation in the dark for 60 days. The amount of $FeCl_2$ added during the anaerobic fermentation biogas formation process was done to reach a level of 1000 mg Fe/L, which has been regarded to be at optimum for CH_4 production.

After rewetting, 49 ml of biogas residue slurry were added on top the soil columns, and mixed into the 2-3 cm surface layer, equivalent to 1½ livestock units. The controls got just the same amount of water. Then the columns were covered with quartz sand to homogenize the added water across the entire surface, and kept in a dark room at 19-21°C. All column experiments were done in triplicate, which made 18 columns in total. 187 eluate solutions have been analyzed, taken within 49 days. Some parameters were measured till day 65.

120 ml of de-ionized water were added 2 times a week on top, which resembles the average natural precipitation of 1000mm on site (inner Alpine area), and the eluates sampled. The evaporation was 1,1-1,2 g per collection vessel. The water content inside the columns was measured by a TDR-probe. The column experiments were performed during 8 weeks to simulate an average growth period of pasture between cuts (Unterfrauner 2009) (Fig 1).

After the experiment, the soil columns were sliced and the profile investigated according to Austrian standard methods, like extractions by de-ionized water (saturation water extract termed as H_2O), 0,4M LiCl (= exchangeables; after Husz 2001), HCl (= supplementaries), and aqua regia (= quasi total). Final determinations were done by ICP-OES and spectrophotometry (ammonium, nitrate). Further fundamental parameters were also done according to Austrian standard methods, like pH, lime contents, humics contents (= organic carbon * 1,72), total N, ammonium, nitrate, and cation exchange capacity (Table 1).

Table1. List of Austrian standard methods used for the analysis of the analysis of the test soils

ÖNORM L1080	Chemical analyses of soils - Determination of organic carbon by dry combustion with and without consideration of carbonates
ÖNORM L1082	Chemical analyses of soils - Determination of nitrogen according to Kjeldahl
ÖNORM L1083	Chemical analyses of soils - Determination of acidity (pH value)
ÖNORM L1084	Chemical analyses of soils - Determination of carbonate
ÖNORM L1085	Chemical analyses of soils - Extraction of elements with aqua regia or with a mixture of nitric- and perchloric-acid
ÖNORM L1086-1	Chemical analyses of soils - Extraction of the effective exchangeable

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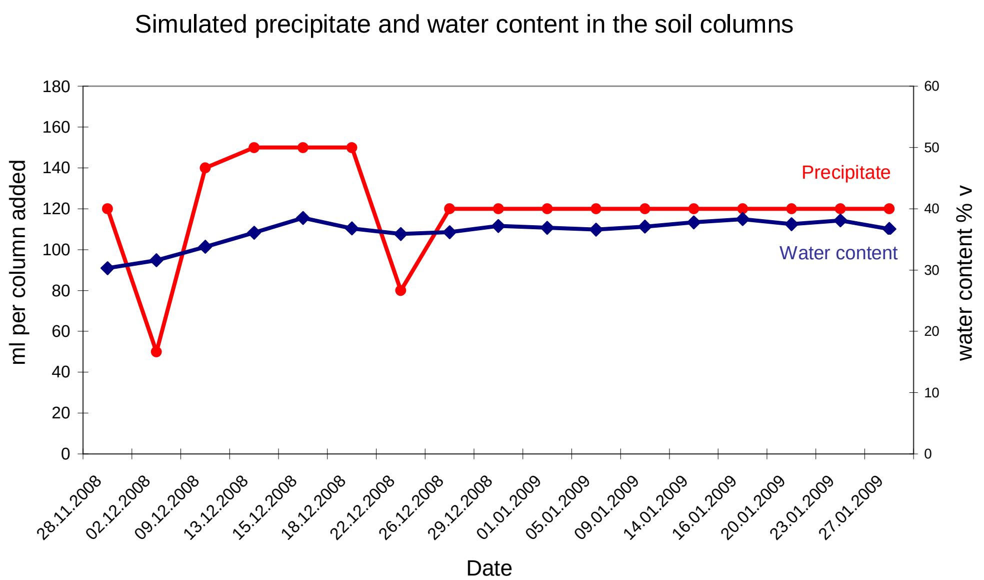
	cations Ca++, K+, Mg++, Na+ and Al+++, Fe+++, Mn++ and H+ by bariumchloride solution and determination of the exchange capacity
ÖNORM L1087	Chemical analysis of soils - Determination of "plant-available" phosphorus and potassium by the calcium-acetate-lactate (CAL)-method
ÖNORM L1092	Chemical analysis of soils - Methods for the extraction of water soluble elements and compounds
ÖNORM L1094-3	Chemical analyses of soils - Extraction of trace elements with a solution of lithium chloride

Fig.1



3.Results and discussion

3.1 Effects of long-term sludge application upon a permanent Alpine grassland soil

The two grassland soils, which have been samples just a few meters apart, but treated in different ways, showed significant differences in fundamental classical parameters (table 2), as well in else mobile fractions and aqua regia (table 3).
Application of urban sewage sludge at 7,5t/ha annually for 10 years, increased organic substance and mobile nutrients in both horizons, due to standard methods of soil analysis (table 2). In particular, increase in water retention capacity, cation exchange capacity, releasable nitrate, sulfate and chloride, and thus increase in electrical conductivity were noted. To the contrary, carbonate and pH (KCl) decreased. The increase of K and P in all mobile soil fractions investigated was paralleled by increases in Ba, Cd, Cr, Cu, Mn, Ni, Pb, V, Zn (in alphabetical order). Data for the A-horizon are given in table 3 and data for the AB- horizon are given in table 4. Changes of mobilities were most pronounced for K, which increased about 100- fold in the saturation water extract, and 18-fold in LiCl extract. The release of

trace elements Cu, Co, Mo, Ni, Cr, and Pb to the water and LiCl- extracts were low, and at the detection limit of the method used. Sludge loading increased the mobilities in LiCl for Zn and Mn more than in water, and increased HCl mobility versus aqua regia for P, K, Cu and Zn. Al, Fe, and Na showed increased mobility, but no increase in aqua regia. The so-called "carbonate-free" cambisol decreased in Ca and Mg instead. Just for Co and Mo, the effects were low. Surprisingly, less than half of P was recovered in HCl, compared with aqua regia, in all cases. The effects were about the same in the A and in the underlaying AB horizon, except for exceptional high increase of Na and Mn release into water from the underlying layer caused by the loading (table 4). With respect to the mobility in dilute HCl, the loaded sample still contained less Zn, Ni and Cr, compared with river sediments of the Danube taken in 1987 (Sager et al. 1990).

Table 2: Changes of fundamental parameters of permanent Alpine grassland by long-term sludge application, prior to manure addition

	A - horizon		AB- horizon		
	grass	**sludge**	**grass**	**sludge**	
Water capacity (ml/100g)	72	84,1	59,1	71,6	increase
pH (H_2O)	7,4	7,5	7,7	7,4	
pH (KCl)	6,6	6,3	7,3	6,2	decrease
% $CaCO_3$	1,6	<0,1	3,8	0,1	decrease
electrical conductivity (mS/cm)	0,321	0,702	0,365	0,619	increase
Organic substance %	3,9	7,2	2,9	5,2	increase
KAKp (cmol/kg)	75	140	58	104	increase
total nitrogen %	0,285	0,426	0,208	0,316	increase
NO_3-N (H_2O) mg/kg	8,4	26,7	7,3	21,4	increase
SO_4 (H_2O) mg/kg	5	22	5	17	increase
Cl (H_2O) mg/kg	2,9	41,2	1,4	40	increase
NH_4-N (H_2O) mg/kg	1,0	1,6	0,4	0,7	increase
NH_4-N (LiCl) mg/kg	20,3	21,1	11	13,7	

pH (H_2O) means the actual soil pH measured in the water extract, whereas pH(KCl) measured after extraction by a KCl-solution indicates a potential pH, which includes exchangeable H^+ versus other cations applied. A difference between pH (H_2O) and pH (KCl) more than one indicates a labile buffer system.

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Table 3: Change of mobilities by long-term sludge application, obtained by batch extraction utilizing saturation water extract, 0,4M LiCl, 2M HCl and aqua regia, for permanent Alpine grassland (A-horizon), given in mg/kg dry weight, prior to manure addition

Element	**H_2O**		**LiCl**		**HCl**		**aqua regia**		**% HCl**	
	grass	sludge	Grass	sludge	grass	sludge	grass	sludge	grass	sludge
Ca	28,2	40,2	971	1077	6711	3975	8155	5090	82,3	78,1
Mg	9,1	12,7	194	222	2717	702	11660	9132	23,3	7,7
Ba	<0,01	<0,01	0,7	1,7	17,9	45,1	33,1	68,7	54,1	65,6
K	0,7	59,9	20,4	339	112	418	1058	1612	10,6	25,9
Na	1,2	7,4	8,0	14,9	21,3	20,5	101,4	92,9	21,0	22,1
P	**1,2**	**3,0**	**18,0**	**36,2**	**476**	**996**	**1137**	**2019**	41,9	49,3
Al	<0,01	<0,01	7,8	15,1	2499	3086	20540	21160	12,2	14,6
Fe	3,4	0,87	12,3	20,5	4538	5047	46770	45170	9,7	11,2
Mn	0,11	0,05	0,62	1,4	396	470	638	725	62,0	64,9
Cu	0,02	0,03	0,13	0,25	10,3	20,9	35,1	53,3	29,4	39,1
Zn	0,03	0,03	0,42	0,96	15,0	63,5	98,2	187	15,3	33,9
Co	<0,01	<0,01	<0,02	<0,02	5,47	6,47	17,9	17,5	30,6	37,0
Mo	<0,01	<0,01	<0,05	<0,05	0,09	0,08	1,04	1,48	8,7	5,4
Ni	<0,01	<0,01	<0,03	<0,04	6,4	8,3	42,6	47,1	15,0	17,6
Cr	<0,01	<0,01	0,07	0,13	4,5	8,2	39,8	58,4	11,3	14,0
Pb	<0,01	<0,01	<0,01	<0,14	0,24	40,3	29	58,9	0,8	68,4
Cd	<0,01	<0,01	<0,01	0,03	0,24	0,43	0,55	0,75	43,6	57,3
V	<0,01	<0,01	0,02	0,06	9,31	11,9	38,6	45,0	24,1	26,4

Table 4: Change of mobilities by long-term sludge application, obtained by batch extraction utilizing saturation water extract, 0,4M LiCl, 2M HCl and aqua regia, for permanent Alpine grassland (AB-horizon), given in mg/kg dry weight, prior to manure addition

Element	**H_2O**		**LiCl**		**HCl**		**Aqua regia**		% HCl	
	grass	sludge	Grass	sludge	Grass	sludge	grass	sludge	grass	sludge
Ca	31	32,3	1001	875	10969	3775	13110	4638	83,7	81,4
Mg	6,2	8,2	107	148	4413	781	13290	9224	33,2	8,5
Ba	<0,01	<0,01	0,5	1,5	18,8	36,6	33,5	56,4	56,1	64,9
K	0,4	41,5	13,8	250	83,8	337	1023	1508	8,2	22,4
Na	0,9	6,9	6,6	14,7	22,4	21,6	107,5	113,1	20,8	19,1
P	**0,33**	**1,17**	**16,0**	**22,5**	**449**	**770**	**986**	**1562**	45,5	49,3
Al	<0,01	<0,01	6,9	9,5	2510	3030	20760	21180	12,1	14,3
Fe	0,55	1,38	7,95	13,3	4110	5428	46520	45530	8,8	11,9
Mn	0,01	0,09	0,33	0,94	403	519	656	756	61,5	68,6
Cu	0,02	0,03	0,2	0,21	9,7	18,4	33,9	48,5	28,6	38,0

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Zn	<0,01	0,03	0,27	0,63	14,0	42,1	92,3	147	15,1	28,7
Co	<0,01	<0,01	<0,02	<0,02	5,42	7,16	17,9	18,1	30,3	39,6
Mo	<0,01	<0,01	<0,04	<0,05	0,17	<0,07	0,86	1,09	19,8	
Ni	<0,01	<0,01	<0,04	<0,03	5,8	7,8	42,5	46	13,6	17,0
Cr	<0,01	<0,01	0,05	0,08	4,75	7,18	40,1	53,1	11,8	13,5
Pb	<0,01	<0,01	<0,12	<0,13	18,6	32,5	33,5	48	55,5	67,7
Cd	<0,01	<0,01	<0,01	0,02	0,25	0,37	0,46	0,69	54,3	53,6
V	<0,01	<0,01	0,02	0,04	8,56	11,9	37,8	44,4	22,6	26,9

3.2 Addition of biogas manure

Table 5 shows the composition of the biogas manure, the mg of amounts added, and the mg of amounts obtained in the eluates after 49 days (mean from 3 columns each). The latter has been calculated as ml eluate times respective concentration, and summed up. After 49 days, a total of 1320 ml water had been added (Fig. 1), which was presumably close to one exchangeable pore volume. The released amounts can be compared with the saturation water extract given in tables 3 and 4, and were expected to be higher from the columns than by batch extraction, like found previously (Sager 2002 and 2004). Differences might appear due to biological activities in the columns during 2 months of ambient temperature. Both soils released more Ca, Mg, and Na from the columns than the respective water extracts obtained from the A and the AB horizons, but clearly less P, Fe, Mn, and Cu. Shaking with 1M LiCl clearly released much more.

Significant differences between the eluate concentrations obtained from untreated grassland soil and the sludge-treated soils mainly for the nutrients nitrate, ammonia, K, P, and Na. Due to the high pH encountered, release of Fe and Mn was marginal. Whereas in the untreated grassland soil (the low nutrient sample), the pH increased and almost approached the value of the manure, it decreased in the high nutrient sample. This also explains changes in release.

Addition of the untreated manure hardly changed the elution from the columns of the untreated grassland. At the sludge-treated soil, manure addition resulted in more K, Na, and P release, and less ammonium (table 5). Addition of the Fe-treated manure released some more sulfur from the untreated grassland soil, probably due to its higher chloride content, as well as more Mg and Ca due to its lower pH(KCl). Addition of the Fe-treated manure to the sludge-treated soil slightly decreased elutions in many cases, maybe because of increased retention capacity caused by Fe-hydroxides (table 6).

Table 5. Addition of biogas manure and effects upon eluted amounts after 49 days

			mg eluted from			
	Manure (% in dry mass)	mg added	grassland soil	grassland soil+manure	sludge treated soil	sludge treated soil + manure
Org. subst	2,0					
NO_3-N	7,74 total N	187	0,205	0,198	23,3	20,3

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NH_4-N			0,020	0,024	0,184	0,167
Ca	22,4	54,1	42,4	45,4	56,9	58,5
Mg	1,02	24,6	22,3	22,4	17,0	8,53
K	5,92	143	0,52	0,37	62,8	75,8
Na	0,61	14,8	1,77	1,73	9,41	11,04
P	1,27	30,5	< 0,005	< 0,005	0,163	0,191
S	0,41	9,8	1,18	1,17	9,16	8,73
Fe	0,37	8,9	0,007	0,009	0,019	0,020
Mn	0,0269	0,65	0,0010	0,0013	0,0045	0,0044
Cu	0,0094	0,23	0,0040	0,0046	0,019	0,024
Zn	*		0,056	0,032	0,018	0,016
B	*		0,017	0,011	0,018	0,024

Table 6. Addition of Fe-treated biogas manure and effects upon eluted amounts after 49 days

			mg eluted			
	Manure (% in dry mass)	mg added	grassland soil	grassland soil +Fe-manure	sludge treated soil	sludge treated soil + Fe-manure
Org. subst	1,9					
NO_3-N	7,74 total N	187	0,205	0,392	23,3	22,5
NH_4-N			0,020	0,038	0,184	0,061
Ca	20,4	49,2	42,4	47,3	56,9	50,9
Mg	0,82	19,7	22,3	29,2	17,0	15,3
K	6,12	148	0,52	0,48	62,8	59,3
Na	1,02	24,6	1,77	2,06	9,41	8,85
P	1,13	27,3	< 0,005	< 0,005	0,158	0,146
S	0,61	14,8	1,18	1,53	9,16	8,61
Fe	2,53	61,0	0,007	0,008	0,016	0,017
Mn	0,0241	0,58	0,0010	0,0022	0,0042	0,0021
Cu	0,0098	0,24	0,0040	0,0056	0,019	0,020
Zn	*	*	0,056	0,032	0,018	0,010
B	*	*	0,017	0,007	0,018	0,016

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Table 7. pH shifts during the column experiments, measured in H_2O and KCl in the sliced solids

manure	horizon	grassland soil	grassland soil+manure	grassland soil +Fe-manure	sludge treated soil	sludge treated soil + manure	sludge treated soil + Fe-manure
pH(H_2O)8,4	A	7,4 →8,1	7,4 →8,1	7,4 →8,1	7,5 →7,3	7,5 →7,1	7,5 →7,0
	AB	7,7 →8,4	7,7 →8,7	7,7 →8,4	7,7 →7,4	7,7 →7,2	7,7 →7,1
pH (KCl)8,1	A	6,6 →7,0	6,6 →7,0	6,6 →7,0	6,3 →6,4	6,3 →6,3	6,3 →6,3
	AB	7,4 →7,1	7,4 →7,2	7,4 →7,2	6,2 →6,5	6,2 →6,5	6,2 →6,4

3.3 Eluates from biogas manure application

Within tables 8-13, the concentrations encountered in the eluates, are shown in detail. As it is not reasonable to calculate a standard deviation from 3 data, the data are given as such to get an impression about the precision obtained.
Some elements in the eluates were largely below detection limits of the ICP-OES (As, Be, Cd, Co, Cr, Mo, Pb, V), but as the current work was focussed on nutrients and contaminants, this was considered sufficient. Data for As, B, Be, Li, Sr, and total sulfur were not done for the solid samples and the manure applied, but in the eluates only.

4. Discussion

Sorption parameters derived from batch experiments are not valid for modelling transport, because in the column, desorption rates limit the exchange of tracers, whereas in batch, mixed kinetics are observed (Isenbeck-Schröter et al. 1993). The average transport velocity of some anions through soils may be larger than that of accompanying water molecules, owing to electrostatic repulsion on negatively charged solid surfaces. This forces the anions into the pore centres, where the velocity is faster, leaving the small capillaries as „anion-excluded volumes“. Anion exclusion increases with increasing cation exchange capacity of the soil matrix. Delayed arrival of Br and Cl was observed in soils and sediments containing significant quantities of variable-charge clay minerals. Whereas in a sandy soil, tritium, chloride and sulfate had the same velocity, in a clay loam from a nearby site, the average velocity increased from tritium to chloride to sulfate, to double. Retardation effects, detected from conductivity measurements, were small for 0,1M solutions, but pronounced for 0,001M solutions of NaCl, $CaCl_2$, Na_2SO_4 and $CaSO_4$ (Seaman, Bertsch, Miller 1995; Gvirtzman, Gorelick 1991; Beese, Van der Ploeg 1979).
Solubilities obtained from batch water extract should be less than solubilities obtained after percolation of the same water volume through the column, because steady flow leads to steady contact with pure solvent, longer contact times, and biological processes (Sager 2004),

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which was found for chernozem soils samples after intense cereal and maize production. This is not necessarily true for grassland soils investigated in this work. Because of the increase of organic carbon, retention sites also increased, and the increase of mobilities of trace elements was less than expected from loading with inorganics only (compare Sager 1991).
The presentation of cumulative eluted amounts in the graphics smoothes experimental fluctuations and permits the addition of trendlines.

4.1 Nitrate and ammonia

Usually, most concern deals with the loss of nitrate and ammonia to deeper soil layers. Fig. 2 and fig. 3 show that significant differences occurred. The output of nitrate from previously high productive and currently bare soils had caused some concerns with respect to high levels in potable waters. This led to recommendations to grow intermediate plant covers after the harvest of cereals and maize (e.g. Stauffer 1993). In the column experiment done within this work, nitrate release from the fertilized sample was significantly above drinking water thresholds. The addition of manure decreased the cumulative output of nitrate from both soil column types, and enhanced losses of ammonia. Fe(II) addition reduced the output of ammonium and increased nitrate output, with respect to the addition of untreated manure. For the non-fertilized sample, there was very low release of NH_4-N, but high uncertainty due to many values below detection limits, whereas from the fertilized sample, there was steady release.

Fig. 2 Cumulative nitrate-N output

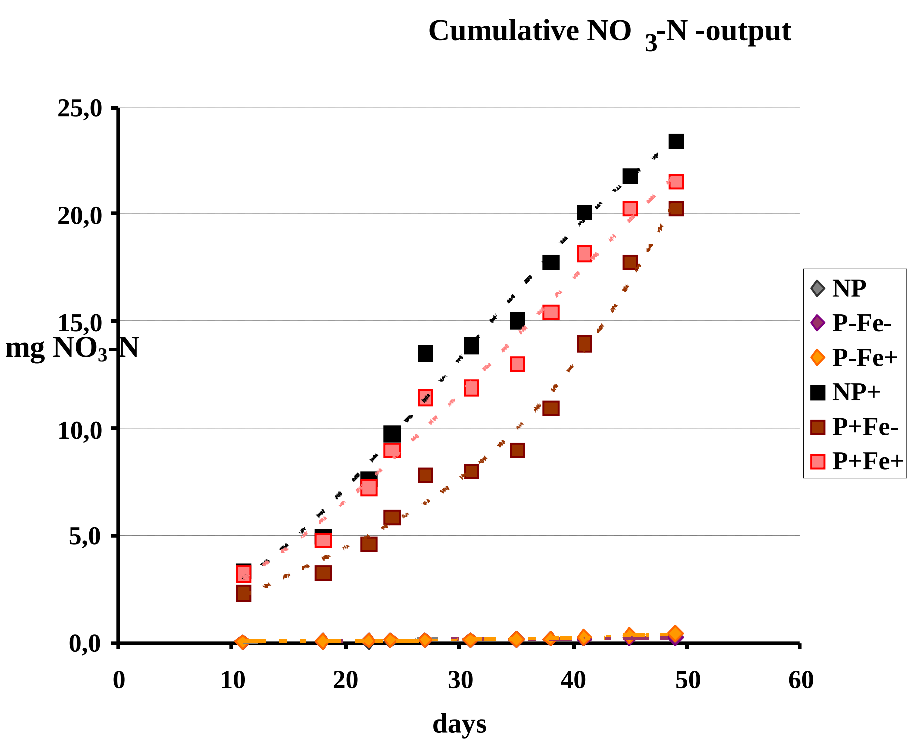


Fig. 3 Cumulative NH_4-N output

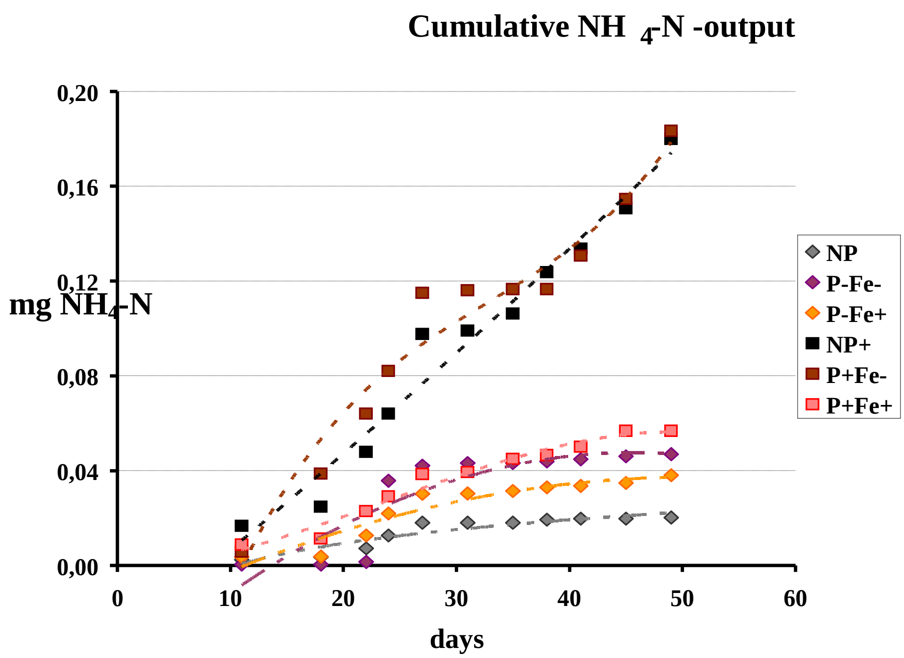
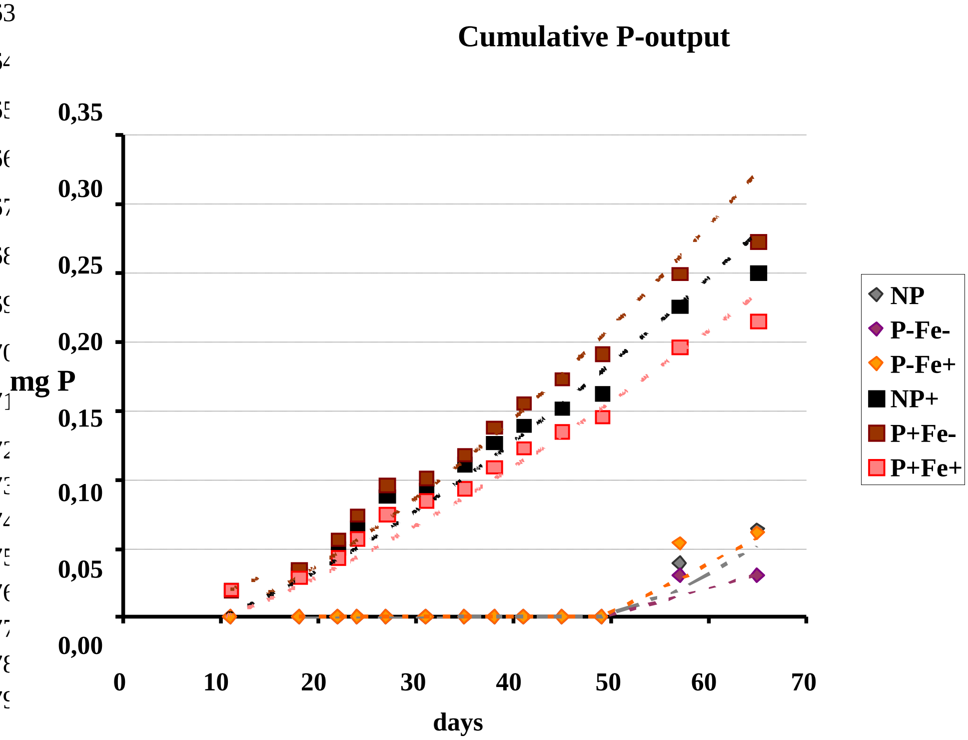


Fig. 4 Cumulative P-output



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4.2 Phosphate

$FeCl_2$ added during the anaerobic fermentation biogas formation process at a level of 1000 mg Fe/L has been regarded to be at optimum for CH_4 production, but its effects on mobile P was low. Similarly, the output from the fertilized soil yielded about constant release of P from the beginning, increased by manure application, and decreased by Fe addition. The release of P from the unfertilized soil was close to detection limits for a long time, and had a lag time of almost one pore volume (fig.4). Usually, P gets liberated from Fe-phosphates during reduction, but anaerobic microorganisms have a large potential of P- uptake from Fe-oxides and storage as well (Fleischer 1986). The re-oxidation of iron sulfides under aerobic conditions might immobilize P, but also lead to acidification.

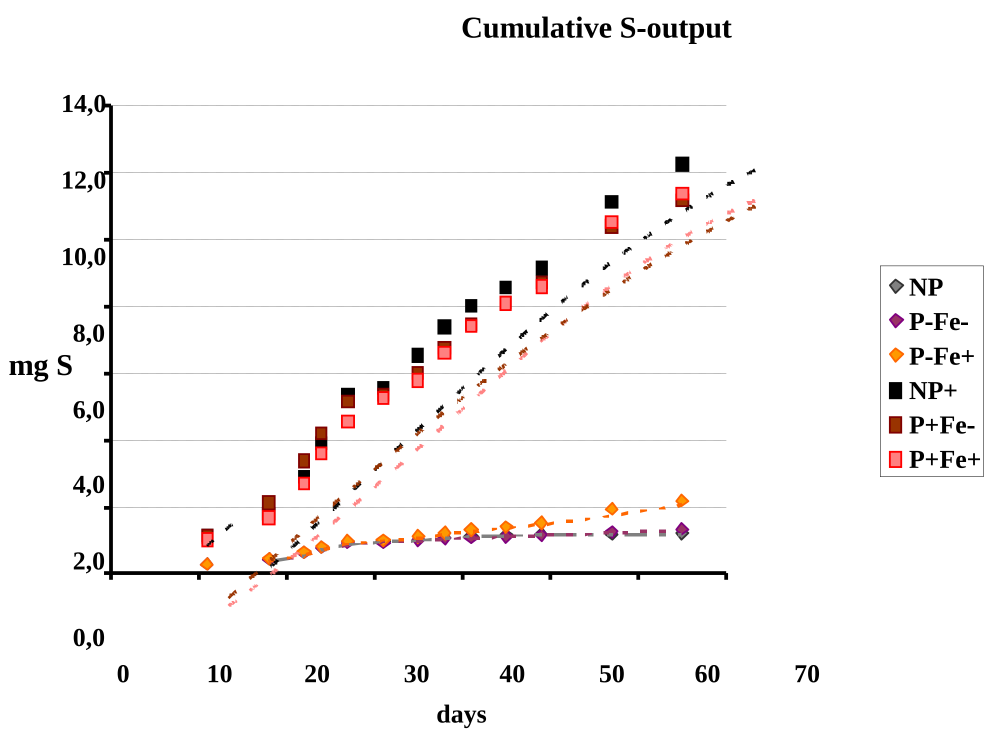
In case phytate has remained from maize additions, it may be important to know that Ca-phytate gets adsorbed at Fe-oxides (goethite) also, but P can be made available by bacteria and plant phytases (Giles et al. 2012).

4.3 Sulfur

pH changes are due to S oxidation and nitrification reactions. Elution of sulfur-compounds from the sludge-pretreated soil columns was significantly higher (fig.5). Slightly lowered sulfur elution can be recognized after manure addition, presumably due to more reducing conditions. Contrary to the assumptions that precipitation of FeS_2 would be effective, the impact of Fe- additions was marginal.

Biogas residues are expected to contain 0,24 – 1,08% S in dry mass (Sager 2007). In our case, the utilized slurry had 95% water, which means max. 0,5 g S per litre, and this should completely react with 1g of Fe(II) per litre.

Fig. 5 Cumulative S-output



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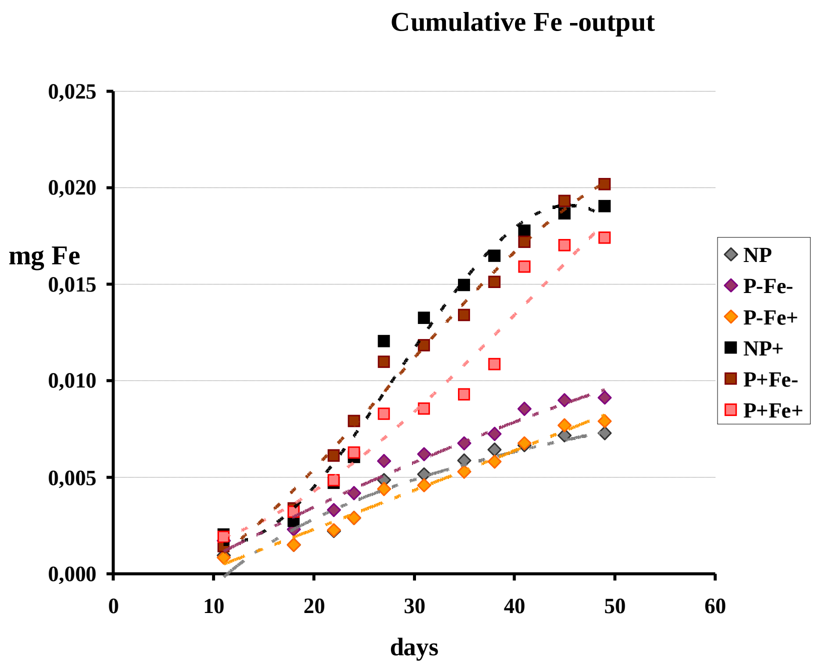
In spite of the high soil pH, some Fe was eluted from both soil column types, more from the sludge pretreated ones in spite of its higher pH. The release of Fe was rather constant, it might migrate either complexed with humics, or divalent. Manure addition slightly lowered the Fe- release (fig. 6). Schärer et al. (2009) investigated the P-sorption of newly precipitated additional iron-hydroxide sorption phases in an Alpine cambisol similar to that used in this experiment, by mixing $Ca(OH)_2$ to the soil, and subsequently $FeSO_4$. Moessbauer spectroscopy revealed the speciation of Fe to be bound as 5-7 % hematite, 25-29% goethite, 10-20% ferrihydrite, phyllosilicates, but no vivianite and pyrite

Fig. 6. Cumulative Fe-output



4.5 Zinc

Zinc was the only element, of which the output from the unfertilized soil was more than from the fertilized one, though it contained less. Contrary to main mutrients, addition of both manure and iron decreased Zn-mobility. Possible explanations are precipitations of ZnS, Zn-phosphates, or increased uptake by microbial biomass (fig.7).

4.6 Copper

From the fertilized soil, release of Cu was much more than from the unfertilized counterpart. Addition of Fe was contradictory, it lowered the release from the fertilized sample, but increased the release from the unfertilized one. Like Zn, Cu might be precipitated as Cu-sulphide, Cu-phosphate, or taken by microbial biomass. The main difference between Cu and Zn is that Cu is bound to humics much more strongly than Zn (fig. 8).

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Fig 7: Cumulative Zn-output

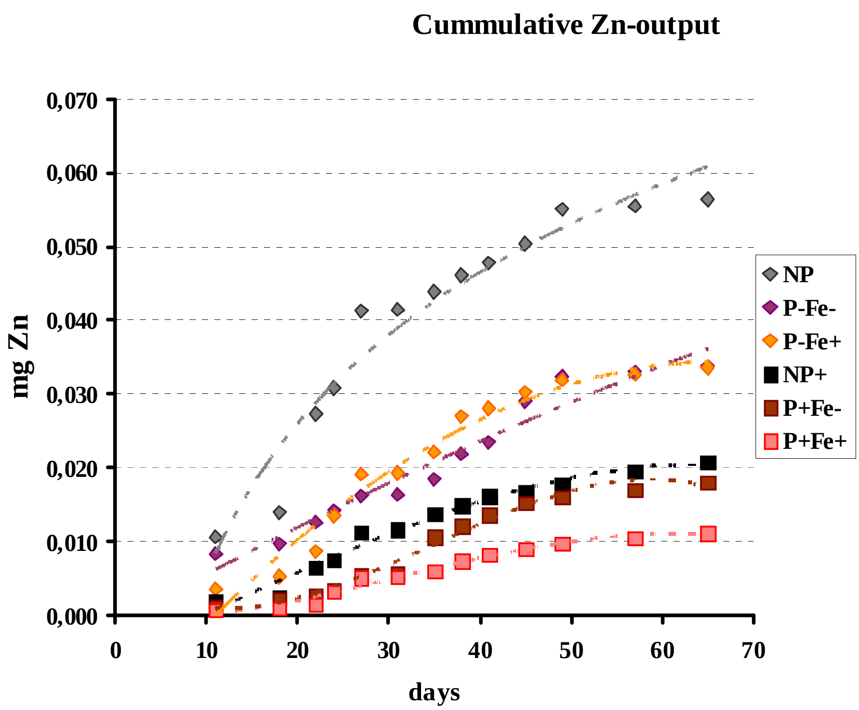
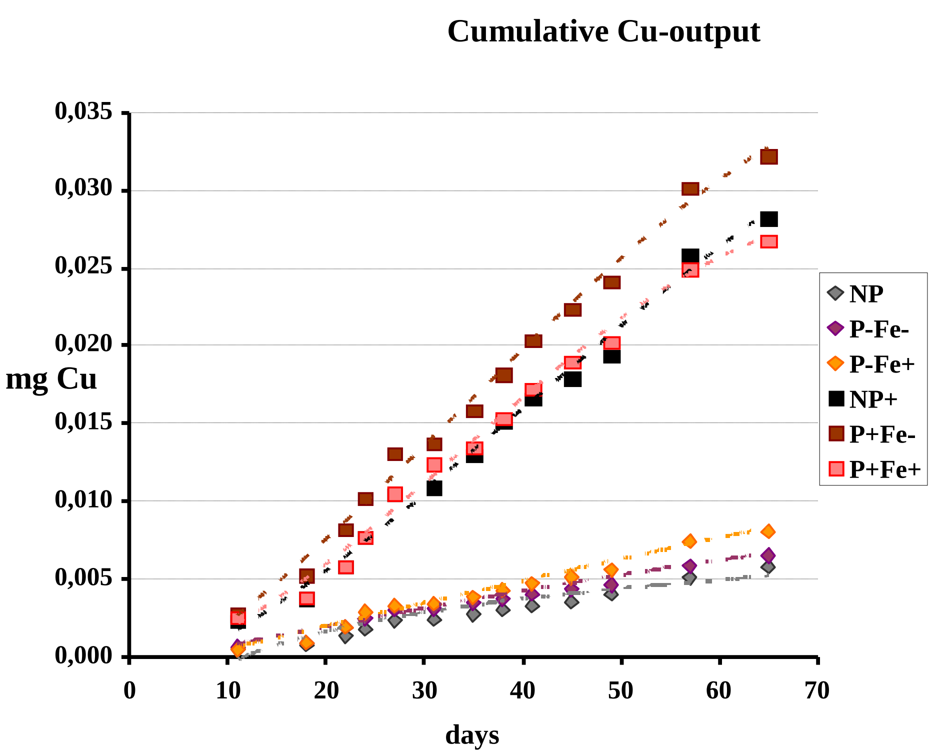


Fig. 8. Cumulative Cu-output



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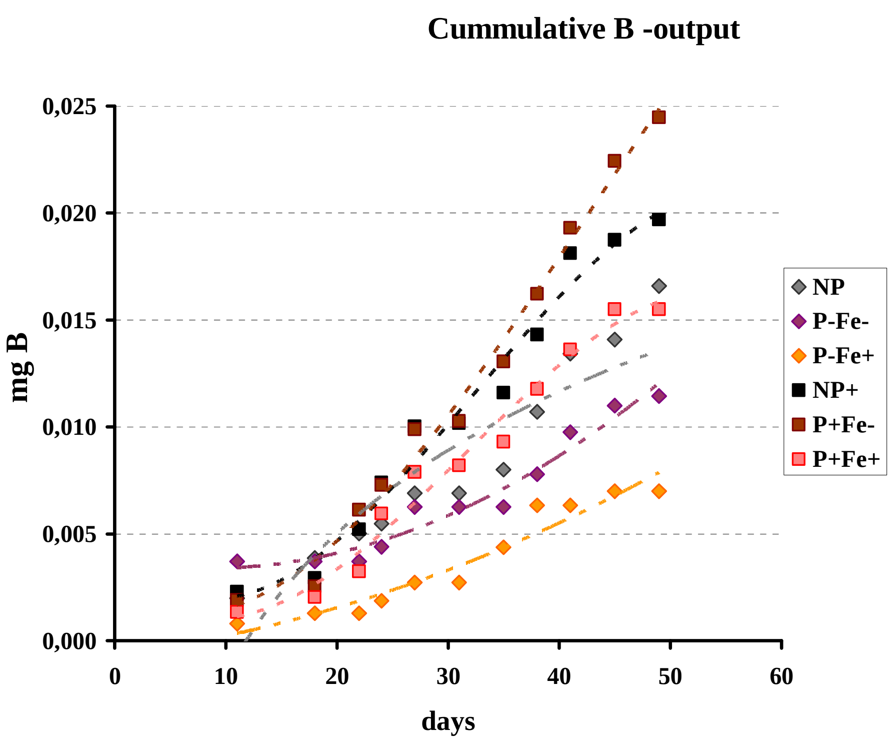
4.7 Boron

With respect to the release of boron, concentration values close or below detection limits had to be considered, and data below detection limit used for fig.9 had been set to zero. Nevertheless, the cummulative output from the fertilized samples was slightly higher, and Fe addition decreased B elution in both cases.

Fig. 9. Cumulative B- output



4.8 Alkalis and alkaline earths

Like the release of K, also K/Na in the eluates was significantly higher from the fertilized sample, but no clear effects of the treatments could be recognized within the observation period, though a lot of Na was applied on top. The cumulative release of Ca was slightly more from the fertilized sample, which can be explained by its lower soil pH (table 7), whereas the effects of the treatments were marginal. The Ca/Mg proportion moved in a constant range of 3,0-3,5 for the fertilized sample, but decreased steadily for the unfertilized one from 3,0-3,5 down to 1,7–1,1; the latter had less Ca/Mg in the solid (tables 5,6). For Mn, the cummulative output was largely negligible, but increased from the fertilized sample within the last 2 fractions.

Explanation for figs 2-9: NP … original grassland soil
P-Fe- … original grassland soil + biogas manure
P-Fe+ … original grassland soil + biogas manure + $FeCl_2$
NP+ … sludge treated soil
P+Fe- … sludge treated soil + biogas manure
P+Fe+ …sludge treated soil + biogas manure + $FeCl_2$

5. Conclusions

If grassland soil has been treated with urban sewage sludge for several years, trace and nutrient mobilities are significantly higher, in spite of higher organic carbon and presumable soil life. Differences between the treatments were larger than differences between the respective soil horizons, appearing for the nutrients N-P-K-S as expected, as well as for Cu, Na, and Sr. There was substitution of acid-mobile Ca and Mg by K and carbon. Nitrate leachates from the nutrient-rich soil clearly superseded the permissible limits for potable water, whereas contents of B-Cr-Cu-Ni-Pb were without concern. Similarly, the respective indicator values were touched for NH_4-N in some cases, whereas the levels of Al, Fe, Mn and Na were much lower. Differences have been shown both for column and for batch experiments, though the mobile phases investigated in this work had been surely optimized for K, and are not ideal for the speciation of Fe, Al and P. Microbial retention did not compensate the saturation of adsorption sites. The retention of solutes contained in manure obtained after biogas evolvation, was not largely influenced by $FeCl_2$-addition, which was intended to fix H_2S as FeS or FeS_2 in the slurry. This counteracted the hypothesis of increased P-retention after $FeCl_2$- addition to biogas residues.

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Further items due to the editors´ manuscript composition guidelines:

7. Team list: none

8. Copyright statement
There are no commercial interests for publishing this text. The author confirms that all analytical data have been done in his lab, and he has designed text and graphs.

9. Code availability
No idea about this

10. Data availability
Primary data have been saved in EXCEL format

13. Author contribution → see 8.

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15. Disclaimer
No idea about this

Figure composition and tables
I am not so firm in computering. I am happy that I have finished this WORD file. The figures are graphs imported to the WORD file. The tables are too large and need landscape formate.

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Table 8. mg/l in the eluates from columns of untreated grassland soil without manure addition

Days	ml	NO_3-N	NH_4-N	**Al**	**As**	**B**	**Ba**	**Be**	**Ca**	**Cd**	**Co**	**Cr**	**Cu**	**Fe**	**K**	**Li**	**Mg**	**Mn**	**Mo**	**Na**	**Ni**	**P**	**Pb**	**S**	**Sr**	**V**	**Zn**
11	99	0,29	<0,03	0,024	<0,02	0,032	0,047	<0,0001	55,9	<0,0005	<0,001	<0,0005	0,006	0,012	0,11	<0,0001	15,1	0,0006	<0,02	1,85	0,003	<0,01	<0,005	3,47	0,063	0,001	0,103
	98	0,32	<0,03	0,011	<0,02	0,028	0,121	<0,0001	39,0	<0,0005	<0,001	<0,0005	0,005	0,012	0,12	0,0001	15,3	0,0009	<0,02	2,12	0,003	<0,01	<0,005	3,52	0,052	0,001	0,180
	64	0,36	0,11	0,021	<0,02	<0,02	0,019	<0,0001	47,0	0,0006	<0,001	<0,0005	0,006	0,007	0,12	0,0002	15,8	0,0005	<0,02	1,20	<0,003	<0,01	<0,005	2,81	0,049	0,001	0,061
18	58	0,36	0,03	0,043	<0,02	0,076	0,207	<0,0001	45,5	<0,0005	<0,001	<0,0005	0,007	0,010	0,18	<0,0001	20,9	0,0010	<0,02	2,44	0,006	<0,01	<0,005	2,85	0,095	<0,001	0,114
	57	0,37	<0,03	0,017	<0,02	0,021	0,042	<0,0001	30,0	<0,0005	<0,001	<0,0005	0,004	0,012	0,16	<0,0001	18,2	0,0008	<0,02	1,97	<0,003	<0,01	<0,005	2,64	0,042	0,001	0,028
	48	0,37	0,03	0,020	<0,02	<0,02	0,032	<0,0001	52,0	<0,0005	<0,001	<0,0005	0,007	0,008	0,64	<0,0001	20,0	0,0007	<0,02	1,75	<0,003	<0,01	<0,005	3,01	0,066	0,001	0,036
22	95	0,17	0,05	0,020	<0,02	0,037	0,138	<0,0001	48,6	<0,0005	<0,001	<0,0005	0,008	0,009	0,09	<0,0001	23,4	0,0009	<0,02	2,09	0,005	<0,01	<0,005	2,18	0,072	0,001	0,183
	108	0,24	0,03	0,012	<0,02	<0,02	0,021	<0,0001	41,6	<0,0005	0,001	<0,0005	0,004	0,007	0,75	0,0001	26,7	0,0010	<0,02	2,00	0,005	<0,01	<0,005	2,24	0,072	<0,001	0,107
	65	0,23	0,05	0,020	<0,02	0,020	0,055	<0,0001	47,7	<0,0005	<0,001	<0,0005	0,006	0,007	0,10	0,0003	22,5	0,0008	<0,02	2,00	<0,003	<0,01	0,006	2,24	0,071	0,001	0,175
24	96	0,24	0,10	0,012	<0,02	<0,02	0,025	<0,0001	100,3	<0,0005	<0,001	<0,0005	0,006	0,008	0,60	0,0002	27,3	0,0023	<0,02	2,20	0,008	<0,01	<0,005	1,88	0,086	0,001	0,019
	82	0,28	<0,03	0,017	<0,02	<0,02	0,041	<0,0001	44,4	<0,0005	<0,001	<0,0005	0,006	0,010	0,97	<0,0001	29,4	0,0025	<0,02	2,05	0,006	<0,01	<0,005	1,73	0,078	0,001	0,050
	68	0,25	0,10	0,018	<0,02	<0,02	0,021	<0,0001	63,1	<0,0005	<0,001	<0,0005	0,004	0,009	0,18	0,0002	27,1	0,0009	<0,02	1,60	<0,003	<0,01	<0,005	1,66	0,072	<0,001	0,069
27	128	0,21	0,04	0,014	<0,02	0,034	0,053	<0,0001	59,5	<0,0005	<0,001	<0,0005	0,005	0,008	0,27	<0,0001	34,4	0,0021	<0,02	2,33	<0,003	<0,01	<0,005	1,38	0,090	0,001	0,103
	133	0,09	0,06	0,010	0,031	<0,02	0,021	<0,0001	70,5	<0,0005	<0,001	<0,0005	0,006	0,005	0,38	0,0001	35,8	0,0011	<0,02	2,25	<0,003	<0,01	<0,005	1,29	0,096	<0,001	0,071
	107	0,27	0,03	0,017	<0,02	0,030	0,054	<0,0001	56,9	<0,0005	<0,001	<0,0005	0,004	0,007	0,30	<0,0001	33,9	0,0014	<0,02	2,29	<0,003	<0,01	<0,005	1,51	0,080	0,001	0,082
31	13	0,25	*	0,057	<0,02	<0,02	0,076	<0,0001	86,8	<0,0005	<0,001	<0,0005	0,006	0,033	0,61	0,0004	24,8	0,0042	<0,02	2,75	0,004	0,02	0,007	1,22	0,090	0,001	0,021
	9	*	<0,2	0,064	<0,02	<0,02	0,033	<0,0001	154,4	<0,0005	<0,001	<0,0005	0,006	0,055	0,39	0,0003	23,3	0,0052	<0,02	2,99	0,007	0,01	<0,005	1,09	0,130	<0,001	0,011
	11	0,14	<0,01	0,022	<0,02	<0,02	0,039	<0,0001	65,4	<0,0005	<0,001	<0,0005	0,005	0,007	1,17	0,0001	30,3	0,0017	<0,02	2,31	0,009	<0,01	<0,005	0,95	0,099	0,001	0,033
35	89	0,18	<0,01	0,008	<0,02	<0,02	0,051	<0,0001	71,3	<0,0005	<0,001	<0,0005	0,005	0,008	0,28	<0,0001	25,4	0,0009	<0,02	2,20	0,003	<0,01	<0,005	0,49	0,094	0,001	0,073
	94	0,12	<0,01	0,005	<0,02	<0,02	0,038	<0,0001	66,4	<0,0005	<0,001	<0,0005	0,005	0,008	0,41	0,0001	32,1	0,0013	<0,02	2,53	<0,003	<0,01	<0,005	1,00	0,099	<0,001	0,052
	58	0,49	<0,05	0,023	<0,02	<0,02	0,032	<0,0001	63,3	<0,0005	<0,001	<0,0005	0,005	0,008	0,79	<0,0001	29,3	0,0011	<0,02	2,48	<0,003	<0,01	<0,005	1,18	0,101	0,001	0,032
38	92	0,27	0,02	0,023	<0,02	<0,02	0,038	<0,0001	75,8	<0,0005	<0,001	<0,0005	0,004	0,011	0,41	<0,0001	36,5	0,0024	<0,02	2,55	0,008	<0,01	<0,005	0,65	0,113	<0,001	0,013
	89	0,47	0,02	0,031	<0,02	0,029	0,065	<0,0001	61,9	<0,0005	<0,001	<0,0005	0,004	0,009	0,46	<0,0001	37,6	0,0011	<0,02	2,69	0,008	<0,01	<0,005	0,67	0,102	0,001	0,022
	64	0,30	<0,01	0,048	<0,02	0,057	0,048	<0,0001	41,3	<0,0005	<0,001	<0,0005	0,003	0,011	0,36	<0,0001	33,9	0,0012	<0,02	2,59	0,007	<0,01	<0,005	0,67	0,078	<0,001	0,057
41	57	0,29	<0,01	0,018	<0,02	<0,02	0,074	<0,0001	46,0	<0,0005	<0,001	<0,0005	0,005	0,008	0,22	<0,0001	29,7	0,0012	<0,02	2,50	<0,003	<0,01	<0,005	0,68	0,099	<0,001	0,057

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	48	0,37	0,02	0,023	<0,02	0,122	0,104	<0,0001	28,6	<0,0005	0,001	<0,0005	0,005	0,008	0,18	<0,0001	32,4	0,0016	<0,02	2,03	<0,003	<0,01	0,005	0,51	0,077	<0,001	0,019
	33	0,39	0,01	0,016	<0,02	0,086	0,035	<0,0001	23,3	<0,0005	<0,001	<0,0005	0,003	0,005	0,21	<0,0001	29,5	0,0012	<0,02	1,93	<0,003	<0,01	0,005	0,47	0,064	<0,001	0,022
45	95	0,34	<0,05	0,010	<0,02	<0,02	0,021	<0,0001	46,6	<0,0005	0,001	<0,0005	0,004	0,006	0,70	0,0001	29,8	0,0010	<0,02	2,21	0,003	<0,01	<0,005	0,56	0,091	<0,001	0,051
	55	0,20	<0,01	0,027	<0,02	0,022	0,041	<0,0001	64,4	<0,0005	<0,001	<0,0005	0,004	0,007	0,59	0,0002	32,5	0,0013	<0,02	2,46	0,006	<0,01	<0,005	0,60	0,117	0,002	0,031
	35	0,41	0,01	0,018	<0,02	0,070	0,031	<0,0001	26,0	<0,0005	<0,001	<0,0005	0,003	0,005	0,24	0,0001	28,9	0,0013	<0,02	1,95	<0,003	<0,01	<0,005	0,33	0,065	0,001	0,034
49	62	0,19	<0,1	0,010	<0,02	0,021	0,039	<0,0001	48,2	<0,0005	<0,001	<0,0005	0,008	0,011	5,16	<0,0001	33,6	0,0013	<0,02	2,44	<0,003	<0,01	<0,005	0,36	0,082	0,001	0,099
	51	0,21	<0,01	0,010	<0,02	0,107	0,883	<0,0001	63,8	<0,0005	<0,001	<0,0005	0,013	0,009	6,08	0,0004	40,2	0,0009	<0,02	5,39	0,004	<0,01	<0,005	0,57	0,127	<0,001	0,133
	43	0,20	<0,1	0,017	<0,02	<0,02	0,034	<0,0001	49,7	<0,0005	<0,001	<0,0005	0,007	0,010	0,84	0,0002	33,3	0,0013	<0,02	2,31	0,005	<0,01	0,005	0,43	0,085	0,002	0,027

* … not determined

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Table 9. mg/l in the eluates from columns of sludge-treated soil without manure addition

Days	ml	NO_3-N	NH_4-N	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Li	Mg	Mn	Mo	Na	Ni	P	Pb	S	Sr	V	Zn
11	87	33,2	0,13	0,029	<0,02	0,026	0,127	<0,0001	74,8	<0,0005	<0,001	0,0009	0,028	0,025	87,8	<0,0001	21,5	0,0012	<0,02	14,1	0,007	0,27	0,005	12,8	0,181	0,003	0,009
	94	40,1	0,29	0,024	<0,02	0,049	0,187	<0,0001	85,3	<0,0005	<0,001	<0,0005	0,024	0,025	91,6	0,0002	25,6	0,0021	<0,02	15,2	0,009	0,24	<0,005	14,2	0,218	0,002	0,047
	75	46,5	0,15	0,020	<0,02	0,033	0,138	<0,0001	85,3	<0,0005	<0,001	0,0008	0,026	0,019	95,6	<0,0001	24,6	0,0023	<0,02	15,3	0,010	0,20	<0,005	14,9	0,209	0,002	0,007
18	51	28,0	0,04	0,036	<0,02	<0,02	0,085	<0,0001	69,4	0,0005	<0,001	<0,0005	0,028	0,021	93,0	<0,0001	24,2	0,0013	<0,02	14,2	0,003	0,24	0,005	9,2	0,122	0,003	0,002
	55	15,9	<0,15	0,116	<0,02	<0,02	0,074	<0,0001	47,3	<0,0005	<0,001	0,0015	0,029	0,096	80,1	0,0002	16,3	0,0060	<0,02	12,1	0,007	0,31	0,005	10,9	0,155	0,003	0,011
	57	31,0	0,39	0,020	<0,02	0,032	0,138	<0,0001	80,4	<0,0005	<0,001	<0,0005	0,026	0,022	94,7	<0,0001	22,5	0,0126	<0,02	15,7	0,006	0,17	<0,005	14,5	0,193	0,002	0,016
22	92	25,0	0,12	0,018	<0,02	<0,02	0,071	<0,0001	69,0	0,0007	<0,001	0,0008	0,019	0,020	54,7	<0,0001	20,2	0,0014	<0,02	7,9	0,007	0,21	<0,005	8,2	0,124	0,002	0,066
	109	30,8	0,25	0,012	<0,02	0,039	0,126	<0,0001	88,4	<0,0005	<0,001	0,0005	0,026	0,020	92,1	0,0001	24,2	0,0016	<0,02	14,6	0,011	0,24	<0,005	14,5	0,222	0,002	0,029
	85	28,7	0,37	0,012	<0,02	<0,02	0,088	<0,0001	81,6	<0,0005	<0,001	<0,0005	0,018	0,019	65,0	<0,0001	25,4	0,0122	<0,02	9,7	0,005	0,14	<0,005	11,6	0,148	0,002	0,037
24	80	25,8	0,08	0,028	<0,02	0,029	0,122	<0,0001	63,9	<0,0005	<0,001	0,0007	0,027	0,021	85,0	<0,0001	21,2	0,0012	<0,02	12,7	0,008	0,23	<0,005	11,3	0,164	0,002	0,014
	82	28,8	0,30	0,012	<0,02	0,031	0,105	<0,0001	67,0	<0,0005	0,002	<0,0005	0,023	0,019	84,5	0,0002	23,4	0,0042	<0,02	13,1	0,009	0,19	<0,005	13,7	0,169	0,002	0,009
	56	33,2	0,31	0,025	<0,02	0,029	0,131	<0,0001	66,7	<0,0005	0,001	0,0007	0,027	0,022	91,7	<0,0001	23,9	0,0258	<0,02	14,7	0,009	0,23	0,006	17,3	0,196	0,002	0,016
27	137	25,5	0,23	0,023	<0,02	0,025	0,106	<0,0001	61,6	<0,0005	<0,001	<0,0005	0,026	0,018	86,8	<0,0001	22,5	0,0016	<0,02	12,5	0,011	0,20	0,007	10,8	0,165	0,002	0,010
	129	32,7	0,23	0,012	0,025	0,034	0,112	<0,0001	72,6	<0,0005	0,001	0,0006	0,024	0,019	89,4	0,0001	24,9	0,0045	<0,02	13,8	0,005	0,19	<0,005	13,8	0,187	0,002	0,010
	114	31,7	0,35	0,009	<0,02	<0,02	0,062	<0,0001	69,7	<0,0005	<0,001	<0,0005	0,015	0,016	49,9	<0,0001	23,9	0,0239	<0,02	7,1	0,006	0,12	<0,005	9,2	0,111	0,001	0,079
31	15	15,9	<0,15	0,116	<0,02	<0,02	0,074	<0,0001	47,3	<0,0005	<0,001	0,0015	0,029	0,096	80,1	0,0002	16,3	0,0060	<0,02	12,1	0,007	0,31	0,005	10,9	0,155	0,003	0,011
	14	16,0	<0,15	0,187	<0,02	0,022	0,110	<0,0001	87,8	<0,0005	0,002	0,0012	0,036	0,155	108,9	0,0003	25,4	0,0390	<0,02	17,4	0,012	0,33	0,007	15,8	0,219	0,003	0,025
	14	16,0	<0,15	0,151	<0,02	0,021	0,092	<0,0001	67,6	<0,0005	<0,001	0,0013	0,032	0,125	94,5	0,0002	20,8	0,0225	<0,02	14,7	0,010	0,32	0,006	13,3	0,187	0,003	0,018
35	92	3,21	0,13	0,018	<0,02	0,025	0,125	<0,0001	63,9	<0,0005	0,002	0,0008	0,027	0,019	85,3	0,0001	18,2	0,0010	<0,02	12,2	0,015	0,25	<0,005	11,5	0,167	0,003	0,043
	83	24,0	0,03	0,015	<0,02	<0,02	0,158	<0,0001	76,1	<0,0005	<0,001	0,0008	0,027	0,020	82,9	0,0001	19,5	0,0011	<0,02	12,8	0,005	0,24	<0,005	13,8	0,188	0,002	0,019
	55	23,9	0,13	0,012	<0,02	<0,02	0,104	<0,0001	65,3	<0,0005	0,002	0,0008	0,030	0,021	82,9	<0,0001	17,1	0,0107	<0,02	12,8	0,008	0,25	<0,005	14,1	0,160	0,003	0,013
38	116	33,7	0,27	0,024	<0,02	0,026	0,088	<0,0001	61,9	<0,0005	<0,001	<0,0005	0,023	0,021	78,2	<0,0001	20,7	0,0014	<0,02	11,3	0,008	0,19	<0,005	9,0	0,153	0,003	0,008
	79	32,2	0,20	0,027	<0,02	0,038	0,117	<0,0001	67,5	<0,0005	<0,001	0,0006	0,024	0,021	72,1	<0,0001	21,6	0,0017	<0,02	10,71	0,009	0,19	<0,005	10,38	0,171	0,003	0,020
	63	28,2	0,08	0,034	<0,02	0,036	0,087	<0,0001	56,5	<0,0005	<0,001	<0,0005	0,026	0,025	72,0	<0,0001	19,3	0,0023	<0,02	10,74	0,006	0,17	<0,005	10,76	0,142	0,003	0,016
41	97	34,5	0,24	0,022	<0,02	0,063	0,114	<0,0001	74,8	<0,0005	<0,001	<0,0005	0,023	0,016	79,2	<0,0001	23,3	0,0018	<0,02	10,7	0,005	0,19	<0,005	8,5	0,169	0,002	0,017

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	63	38,0	0,08	0,035	<0,02	0,044	0,101	<0,0001	62,4	<0,0005	<0,001	0,0006	0,024	0,019	74,5	0,0001	22,3	0,0013	<0,02	11,06	0,007	0,16	<0,005	10,53	0,157	0,003	0,027
	41	30,0	0,04	0,043	<0,02	0,035	0,089	<0,0001	61,6	<0,0005	0,001	0,0006	0,024	0,027	68,7	<0,0001	18,7	0,0024	<0,02	11,81	0,005	0,17	<0,005	10,04	0,141	0,002	0,016
45	96	34,8	0,53	0,016	<0,02	<0,02	0,120	<0,0001	78,6	<0,0005	<0,001	0,0006	0,023	0,018	94,8	0,0001	21,9	0,0045	<0,02	12,5	0,009	0,24	<0,005	9,6	0,204	0,003	0,007
	49	16,1	<0,05	0,013	<0,02	<0,02	0,097	<0,0001	64,7	<0,0005	0,001	0,0010	0,022	0,017	76,1	0,0001	18,1	0,0013	<0,02	11,4	0,013	0,20	<0,005	10,7	0,163	0,002	0,013
	30	29,8	0,02	0,025	<0,02	0,061	0,087	<0,0001	56,8	<0,0005	<0,001	<0,0005	0,023	0,017	64,5	<0,0001	17,2	0,0017	<0,02	9,6	0,006	0,17	<0,005	8,9	0,129	0,002	0,019
49	75	24,1	1,17	0,022	<0,02	<0,02	0,102	<0,0001	84,9	<0,0005	<0,001	0,0005	0,024	0,018	98,6	<0,0001	25,8	0,0152	<0,02	12,4	<0,003	0,22	0,006	9,0	0,203	0,003	0,007
	64	39,7	<0,1	0,027	<0,02	<0,02	0,155	<0,0001	96,0	<0,0005	0,001	0,0005	0,025	0,019	86,4	0,0002	21,1	0,0084	<0,02	12,9	0,012	0,20	<0,005	10,8	0,201	0,002	0,033
	24	20,2	<0,15	0,038	<0,02	<0,02	0,095	<0,0001	59,0	<0,0005	0,001	0,0005	0,029	0,029	89,5	<0,0001	18,4	0,0091	<0,02	13,1	0,010	0,18	<0,005	13,0	0,155	0,002	0,013

* … not determined

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Table 10. mg/l in the eluates from columns of original grassland soil after manure addition

Days	ml	NO_3-N	NH_4-N	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Li	Mg	Mn	Mo	Na	Ni	P	Pb	S	Sr	V	Zn
11	96	0,20	0,01	0,040	<0,02	0,022	0,048	<0,0001	50,0	<0,0005	<0,001	<0,0005	0,006	0,011	0,64	<0,0001	16,8	0,0010	<0,02	1,59	0,007	<0,01	<0,005	3,05	0,066	<0,001	0,028
	115	0,25	<0,03	0,035	<0,02	0,078	0,191	<0,0001	56,8	<0,0005	<0,001	<0,0005	0,009	0,031	0,13	<0,0001	14,6	0,0008	<0,02	2,35	0,003	0,01	<0,005	2,69	0,069	0,001	0,187
	75	0,38	<0,01	0,024	<0,02	<0,02	0,026	<0,0001	46,2	<0,0005	<0,001	<0,0005	0,006	0,008	0,32	<0,0001	16,0	0,0006	<0,02	1,64	<0,003	<0,01	<0,005	3,75	0,061	<0,001	0,014
18	60	0,29	<0,03	0,016	<0,02	<0,02	0,042	<0,0001	44,8	<0,0005	<0,001	<0,0005	0,006	0,010	0,15	<0,0001	19,5	0,0007	<0,02	1,96	0,005	<0,01	<0,005	2,52	0,072	0,001	0,045
	56	0,35	<0,03	0,017	<0,02	<0,02	0,025	<0,0001	44,7	<0,0005	<0,001	<0,0005	0,005	0,013	0,34	0,0002	16,3	0,0008	<0,02	1,67	<0,003	<0,01	<0,005	2,01	0,058	0,001	0,012
	49	0,48	<0,02	0,026	<0,02	<0,02	0,029	<0,0001	56,8	<0,0005	<0,001	<0,0005	0,004	0,015	0,15	0,0001	18,0	0,0009	<0,02	1,89	<0,003	<0,01	<0,005	3,22	0,072	0,002	0,009
22	109	0,21	0,03	0,013	<0,02	<0,02	0,024	<0,0001	48,3	<0,0005	0,001	<0,0005	0,021	0,009	0,42	0,0002	24,5	0,0010	<0,02	2,01	0,005	<0,01	<0,005	1,96	0,083	0,001	0,047
	110	0,17	<0,01	0,023	<0,02	<0,02	0,023	<0,0001	50,8	<0,0005	<0,001	<0,0005	0,004	0,008	0,63	<0,0001	23,1	0,0009	<0,02	1,73	<0,003	<0,01	<0,005	1,47	0,078	<0,001	0,016
	80	0,41	*	0,026	<0,02	<0,02	0,029	<0,0001	44,0	<0,0005	<0,001	<0,0005	0,004	0,014	0,32	0,0002	23,0	0,0011	<0,02	2,06	0,004	<0,01	<0,005	2,71	0,083	0,002	0,026
24	100	0,23	0,95	0,027	<0,02	0,023	0,039	<0,0001	62,5	<0,0005	<0,001	<0,0005	0,007	0,009	0,41	0,0001	23,1	0,0014	<0,02	1,90	<0,003	<0,01	<0,005	1,11	0,088	0,001	0,025
	92	0,31	0,17	0,014	<0,02	<0,02	0,029	<0,0001	55,4	<0,0005	0,002	<0,0005	0,005	0,007	0,29	0,0002	28,0	0,0013	<0,02	2,33	<0,003	<0,01	<0,005	2,35	0,086	0,001	0,011
27	116	0,08	0,06	0,014	<0,02	0,023	0,019	<0,0001	56,5	0,0006	<0,001	<0,0005	0,006	0,007	0,63	0,0003	32,9	0,0017	<0,02	2,35	<0,003	<0,01	0,012	1,44	0,096	0,001	0,003
	127	0,14	0,08	0,021	<0,02	<0,02	0,024	<0,0001	71,0	<0,0005	<0,001	<0,0005	0,005	0,009	0,68	<0,0001	26,5	0,0017	<0,02	2,11	<0,003	<0,01	<0,005	0,78	0,108	0,001	0,011
	99	0,20	0,02	0,015	0,027	<0,02	0,028	<0,0001	64,7	<0,0005	0,001	<0,0005	0,005	0,008	0,40	0,0001	34,2	0,0017	<0,02	2,34	0,004	<0,01	<0,005	1,96	0,100	<0,001	0,042
31	9	0,26	0,22	0,043	<0,02	<0,02	0,070	<0,0001	97,9	<0,0005	<0,001	<0,0005	0,005	0,028	0,75	0,0002	23,9	0,0051	<0,02	2,81	<0,003	<0,01	<0,005	1,17	0,131	<0,001	0,020
	8	*	*	0,105	<0,02	<0,02	0,053	<0,0001	*	<0,0005	<0,001	<0,0005	0,006	0,080	1,44	0,0002	19,0	0,0165	<0,02	2,44	0,005	0,02	0,007	0,69	0,113	<0,001	0,024
35	96	0,14	<0,01	0,015	<0,02	<0,02	0,049	<0,0001	63,6	<0,0005	<0,001	<0,0005	0,005	0,007	0,89	<0,0001	31,5	0,0012	<0,02	2,53	0,013	<0,01	<0,005	0,91	0,099	0,001	0,020
	80	0,29	<0,01	0,011	0,020	<0,02	0,022	<0,0001	46,8	<0,0005	<0,001	<0,0005	0,004	0,006	0,45	0,0002	27,9	0,0010	<0,02	2,39	0,005	<0,01	<0,005	1,63	0,087	0,002	0,038
38	91	0,25	0,02	0,021	<0,02	0,029	0,023	<0,0001	50,0	<0,0005	<0,001	<0,0005	0,003	0,008	0,79	0,0002	38,5	0,0017	<0,02	2,05	<0,003	<0,01	<0,005	0,60	0,089	0,002	0,055
	71	0,33	<0,01	0,016	<0,02	0,028	0,046	<0,0001	56,9	<0,0005	0,002	<0,0005	0,004	0,008	0,22	<0,0001	28,8	0,0016	<0,02	2,23	0,006	<0,01	<0,005	0,18	0,088	0,002	0,069
	60	0,32	<0,02	0,026	<0,02	<0,02	0,033	<0,0001	44,2	<0,0005	<0,001	<0,0005	0,004	0,007	0,12	0,0001	31,4	0,0013	<0,02	2,08	0,004	<0,01	<0,005	1,23	0,099	0,001	0,011
41	100	0,26	0,03	0,014	0,022	0,027	0,025	<0,0001	55,3	<0,0005	0,001	<0,0005	0,004	0,008	0,55	<0,0001	42,1	0,0019	<0,02	2,17	<0,003	<0,01	<0,005	0,54	0,101	<0,001	0,031
	65	0,35	<0,02	0,028	<0,02	0,028	0,026	<0,0001	44,7	<0,0005	<0,001	<0,0005	0,005	0,007	0,11	<0,0001	28,8	0,0014	<0,02	1,62	<0,003	<0,01	<0,005	0,12	0,085	<0,001	0,020
	48	0,56	<0,01	0,017	<0,02	<0,02	0,023	<0,0001	29,6	<0,0005	0,001	<0,0005	0,005	0,005	0,35	<0,0001	30,6	0,0012	<0,02	1,93	0,068	<0,01	<0,005	1,017	0,074	<0,001	0,009

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45	117	0,14	0,03	0,007	<0,02	0,020	0,127	<0,0001	97,0	<0,0005	<0,001	<0,0005	0,005	0,015	0,36	0,0003	41,4	0,0023	<0,02	2,99	0,007	<0,01	<0,005	0,89	0,141	0,001	0,126
	51	0,42	<0,01	0,017	<0,02	0,023	0,033	<0,0001	48,8	<0,0005	<0,001	<0,0005	0,004	0,008	0,23	<0,0001	28,9	0,0014	<0,02	2,10	0,006	<0,01	<0,005	0,16	0,084	0,001	0,028
	13	0,52	<0,01	0,034	<0,02	0,028	0,063	<0,0001	25,9	<0,0005	0,001	<0,0005	0,003	0,005	0,18	<0,0001	22,0	0,0012	<0,02	2,00	0,012	<0,01	<0,005	0,83	0,061	<0,001	0,020
49	91	0,14	0,03	0,009	0,022	<0,02	0,038	<0,0001	78,6	<0,0005	<0,001	<0,0005	0,005	0,009	1,06	0,0001	44,4	0,0081	<0,02	2,82	<0,003	<0,01	<0,005	0,83	0,139	0,002	0,075
	47	0,24	<0,01	0,012	<0,02	<0,02	0,042	<0,0001	66,2	<0,0005	<0,001	<0,0005	0,007	0,009	1,19	<0,0001	34,8	0,0011	<0,02	2,28	0,007	<0,01	0,006	0,26	0,119	0,001	0,068
	13	*	<0,3	0,056	<0,02	<0,02	0,089	<0,0001	74,5	<0,0005	<0,001	<0,0005	0,008	0,039	0,64	0,0004	30,1	0,0068	<0,02	2,89	0,003	<0,01	<0,005	1,33	0,091	<0,001	0,019

* … not determined

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Table 11. mg/l in the eluates from columns of sludge-treated soil after manure addition

Days	ml	NO3-N	NH4-N	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Li	Mg	Mn	Mo	Na	Ni	P	Pb	S	Sr	V	Zn
11	101	*	*	0,044	<0,02	<0,02	0,062	<0,0001	55,2	<0,0005	<0,001	0,0007	0,028	0,038	75,4	0,0001	14,1	0,0195	<0,02	11,9	0,009	0,31	0,005	7,0	0,135	0,002	0,009
	80	20,3	0,19	0,045	<0,02	0,044	0,089	0,0001	54,7	<0,0005	0,002	0,0007	0,031	0,033	81,1	<0,0001	16,5	0,0022	<0,02	12,6	0,006	0,23	<0,005	15,5	0,151	0,003	0,009
	75	39,9	0,03	0,027	<0,02	0,033	0,146	<0,0001	101,3	<0,0005	<0,001	0,0005	0,038	0,023	94,6	0,0001	25,2	0,0014	<0,02	15,5	0,008	0,26	<0,005	14,0	0,245	0,002	0,022
18	130	2,93	0,71	0,036	0,022	<0,02	0,110	<0,0001	50,8	<0,0005	<0,001	<0,0005	0,032	0,020	86,4	<0,0001	20,2	0,0153	<0,02	13,0	0,009	0,20	0,005	11,9	0,155	0,003	0,012
	56	9,32	0,08	0,047	<0,02	<0,02	0,093	<0,0001	44,0	0,0006	<0,001	<0,0005	0,037	0,034	89,2	<0,0001	17,0	0,0019	<0,02	13,8	0,007	0,26	<0,005	11,1	0,137	0,003	0,004
	57	34,7	0,03	0,033	<0,02	0,034	0,104	<0,0001	71,1	<0,0005	0,002	0,0007	0,023	0,021	93,1	0,0001	23,2	0,0032	<0,02	15,0	0,010	0,16	<0,005	14,5	0,195	0,002	0,018
22	108	9,65	0,46	0,033	<0,02	0,044	0,080	<0,0001	52,4	<0,0005	0,001	<0,0005	0,029	0,025	88,9	<0,0001	18,9	0,0039	<0,02	12,7	0,010	0,21	0,008	9,6	0,155	0,003	0,003
	101	4,90	0,17	0,047	0,020	0,033	0,084	<0,0001	50,5	<0,0005	0,002	0,0007	0,035	0,039	84,6	0,0003	16,3	0,0019	<0,02	12,7	0,009	0,26	0,007	14,3	0,149	0,003	0,004
	77	32,3	0,12	0,020	<0,02	0,026	0,119	<0,0001	75,2	<0,0005	<0,001	0,0005	0,027	0,021	97,1	0,0002	24,6	0,0023	<0,02	15,6	0,011	0,19	<0,005	16,7	0,187	0,003	0,015
24	79	11,79	0,48	0,027	<0,02	<0,02	0,098	<0,0001	55,8	<0,0005	<0,001	<0,0005	0,029	0,025	81,6	0,0001	18,4	0,0078	<0,02	12,3	0,013	0,25	<0,005	8,0	0,141	0,003	0,008
	84	4,95	0,17	0,035	<0,02	0,022	0,083	<0,0001	51,8	<0,0005	<0,001	0,0007	0,028	0,028	75,6	<0,0001	17,4	0,0016	<0,02	11,1	0,011	0,23	0,005	11,5	0,136	0,002	0,014
	64	36,3	0,03	0,021	<0,02	0,021	0,101	<0,0001	74,0	<0,0005	<0,001	0,0004	0,022	0,019	89,3	<0,0001	22,6	0,0016	<0,02	14,0	0,010	0,21	<0,005	13,2	0,190	0,002	0,007
27	117	9,35	0,35	0,025	<0,02	<0,02	0,083	<0,0001	48,1	<0,0005	<0,001	0,0006	0,025	0,021	84,0	<0,0001	21,0	0,0065	<0,02	11,6	0,011	0,20	<0,005	6,7	0,127	0,002	0,008
	113	11,9	0,41	0,027	<0,02	0,052	0,141	<0,0001	54,5	<0,0005	<0,001	0,0006	0,029	0,026	81,2	<0,0001	18,6	0,0019	<0,02	11,9	0,011	0,22	0,005	10,4	0,148	0,004	0,042
	95	36,6	0,12	0,031	<0,02	<0,02	0,097	<0,0001	45,9	<0,0005	<0,001	0,0004	0,025	0,016	98,8	<0,0001	19,7	0,0011	<0,02	14,7	0,005	0,19	0,005	9,6	0,126	0,002	0,002
35	92	3,53	<0,04	0,027	<0,02	0,056	0,465	<0,0001	51,6	<0,0005	<0,001	0,0007	0,030	0,021	78,5	0,0002	16,4	0,0015	<0,02	12,8	0,018	0,24	<0,005	6,3	0,139	0,002	0,118
	97	10,1	<0,01	0,018	<0,02	0,023	0,092	<0,0001	50,8	<0,0005	0,002	<0,0005	0,028	0,019	82,4	0,0002	16,7	0,0010	<0,02	11,6	0,018	0,21	<0,005	9,0	0,136	0,002	0,035
	40	40,6	0,04	0,018	<0,02	<0,02	0,116	<0,0001	70,2	<0,0005	<0,001	0,0009	0,028	0,022	87,6	<0,0001	18,2	0,0020	<0,02	13,3	0,007	0,24	<0,005	14,0	0,176	0,002	0,021
38	108	17,35	<0,02	0,034	<0,02	0,024	0,079	<0,0001	55,5	<0,0005	0,001	0,0006	0,023	0,021	72,6	0,0001	20,0	0,0019	<0,02	9,64	0,013	0,20	<0,005	5,3	0,143	0,003	0,015
	102	13,54	<0,02	0,036	<0,02	0,025	0,080	<0,0001	58,1	<0,0005	<0,001	<0,0005	0,025	0,021	74,4	0,0001	19,6	0,0017	<0,02	10,23	0,010	0,18	<0,005	7,3	0,145	0,003	0,013
	84	33,3	<0,02	0,040	<0,02	0,025	0,111	<0,0001	72,7	<0,0005	<0,001	<0,0005	0,023	0,021	78,1	<0,0001	21,8	0,0015	<0,02	11,70	0,008	0,22	<0,005	11,4	0,176	0,003	0,017
41	98	27,5	0,15	0,018	<0,02	0,026	0,089	<0,0001	76,0	<0,0005	<0,001	<0,0005	0,021	0,017	78,8	<0,0001	23,9	0,0037	<0,02	10,0	0,006	0,18	<0,005	5,0	0,165	0,002	0,005
	102	28,7	0,20	0,024	<0,02	0,031	0,090	<0,0001	70,6	<0,0005	<0,001	<0,0005	0,023	0,016	77,2	<0,0001	22,5	0,0029	<0,02	10,1	0,006	0,18	<0,005	6,3	0,157	0,003	0,014
	93	36,0	0,08	0,029	<0,02	0,050	0,131	<0,0001	70,0	<0,0005	<0,001	<0,0005	0,023	0,019	76,3	<0,0001	23,2	0,0017	<0,02	11,17	0,009	0,16	<0,005	10,3	0,177	0,002	0,030

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45	105	32,7	0,29	0,038	<0,02	0,038	0,207	<0,0001	70,5	<0,0005	<0,001	<0,0005	0,019	0,018	74,8	<0,0001	22,8	0,0178	<0,02	10,30	0,014	0,18	<0,005	4,8	0,172	0,002	0,023
	100	38,9	0,24	0,027	<0,02	<0,02	0,133	<0,0001	79,1	<0,0005	0,002	<0,0005	0,021	0,019	79,8	<0,0001	23,4	0,0092	<0,02	11,11	0,010	0,18	<0,005	6,2	0,184	0,003	0,008
	92	44,4	0,19	0,035	<0,02	0,039	0,132	<0,0001	77,4	<0,0005	<0,001	<0,0005	0,021	0,020	77,9	<0,0001	24,3	0,0024	<0,02	12,32	0,012	0,19	<0,005	9,4	0,188	0,002	0,017
49	95	23,8	0,09	0,015	<0,02	<0,02	0,124	<0,0001	93,8	<0,0005	<0,001	<0,0005	0,023	0,021	98,7	<0,0001	26,5	0,0300	<0,02	12,5	0,005	0,20	<0,005	6,9	0,215	0,003	0,008
	79	23,3	0,83	0,027	<0,02	0,023	0,102	<0,0001	79,7	<0,0005	<0,001	<0,0005	0,021	0,027	87,8	0,0002	21,8	0,0010	<0,02	10,5	0,007	0,24	<0,005	5,4	0,204	0,003	0,006
	68	50,2	<0,1	0,031	<0,02	0,032	0,132	<0,0001	83,9	<0,0005	<0,001	0,0006	0,022	0,026	91,5	<0,0001	23,9	0,0049	<0,02	13,1	0,004	0,22	0,007	9,7	0,206	0,002	0,019

* … not determined

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Table 12. mg/l in the eluates from columns of original grassland soil after addition of Fe-treated manure

Days	ml	NO_3-N	NH_4-N	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Li	Mg	Mn	Mo	Na	Ni	P	Pb	S	Sr	V	Zn
11	100	*	*	0,044	<0,02	<0,02	0,062	<0,0001	55,2	<0,0005	<0,001	0,0007	0,028	0,038	75,4	0,0001	14,1	0,0195	<0,02	11,9	0,009	0,31	0,005	7,0	0,135	0,002	0,009
	91	20,3	0,19	0,045	<0,02	0,044	0,089	0,0001	54,7	<0,0005	0,002	0,0007	0,031	0,033	81,1	<0,0001	16,5	0,0022	<0,02	12,6	0,006	0,23	<0,005	15,5	0,151	0,003	0,009
	64	39,9	0,03	0,027	<0,02	0,033	0,146	<0,0001	101,3	<0,0005	<0,001	0,0005	0,038	0,023	94,6	0,0001	25,2	0,0014	<0,02	15,5	0,008	0,26	<0,005	14,0	0,245	0,002	0,022
18	64	2,93	0,71	0,036	0,022	<0,02	0,110	<0,0001	50,8	<0,0005	<0,001	<0,0005	0,032	0,020	86,4	<0,0001	20,2	0,0153	<0,02	13,0	0,009	0,20	0,005	11,9	0,155	0,003	0,012
	55	9,32	0,08	0,047	<0,02	<0,02	0,093	<0,0001	44,0	0,0006	<0,001	<0,0005	0,037	0,034	89,2	<0,0001	17,0	0,0019	<0,02	13,8	0,007	0,26	<0,005	11,1	0,137	0,003	0,004
	53	34,7	0,03	0,033	<0,02	0,034	0,104	<0,0001	71,1	<0,0005	0,002	0,0007	0,023	0,021	93,1	0,0001	23,2	0,0032	<0,02	15,0	0,010	0,16	<0,005	14,5	0,195	0,002	0,018
22	85	9,65	0,46	0,033	<0,02	0,044	0,080	<0,0001	52,4	<0,0005	0,001	<0,0005	0,029	0,025	88,9	<0,0001	18,9	0,0039	<0,02	12,7	0,010	0,21	0,008	9,6	0,155	0,003	0,003
	97	4,90	0,17	0,047	0,020	0,033	0,084	<0,0001	50,5	<0,0005	0,002	0,0007	0,035	0,039	84,6	0,0003	16,3	0,0019	<0,02	12,7	0,009	0,26	0,007	14,3	0,149	0,003	0,004
	78	32,3	0,12	0,020	<0,02	0,026	0,119	<0,0001	75,2	<0,0005	<0,001	0,0005	0,027	0,021	97,1	0,0002	24,6	0,0023	<0,02	15,6	0,011	0,19	<0,005	16,7	0,187	0,003	0,015
24	85	11,79	0,48	0,027	<0,02	<0,02	0,098	<0,0001	55,8	<0,0005	<0,001	<0,0005	0,029	0,025	81,6	0,0001	18,4	0,0078	<0,02	12,3	0,013	0,25	<0,005	8,0	0,141	0,003	0,008
	83	4,95	0,17	0,035	<0,02	0,022	0,083	<0,0001	51,8	<0,0005	<0,001	0,0007	0,028	0,028	75,6	<0,0001	17,4	0,0016	<0,02	11,1	0,011	0,23	0,005	11,5	0,136	0,002	0,014
	78	36,3	0,03	0,021	<0,02	0,021	0,101	<0,0001	74,0	<0,0005	<0,001	0,0004	0,022	0,019	89,3	<0,0001	22,6	0,0016	<0,02	14,0	0,010	0,21	<0,005	13,2	0,190	0,002	0,007
27	122	9,35	0,35	0,025	<0,02	<0,02	0,083	<0,0001	48,1	<0,0005	<0,001	0,0006	0,025	0,021	84,0	<0,0001	21,0	0,0065	<0,02	11,6	0,011	0,20	<0,005	6,7	0,127	0,002	0,008
	115	11,9	0,41	0,027	<0,02	0,052	0,141	<0,0001	54,5	<0,0005	<0,001	0,0006	0,029	0,026	81,2	<0,0001	18,6	0,0019	<0,02	11,9	0,011	0,22	0,005	10,4	0,148	0,004	0,042
	104	36,6	0,12	0,031	<0,02	<0,02	0,097	<0,0001	45,9	<0,0005	<0,001	0,0004	0,025	0,016	98,8	<0,0001	19,7	0,0011	<0,02	14,7	0,005	0,19	0,005	9,6	0,126	0,002	0,002
35	101	3,53	<0,04	0,027	<0,02	0,056	0,465	<0,0001	51,6	<0,0005	<0,001	0,0007	0,030	0,021	78,5	0,0002	16,4	0,0015	<0,02	12,8	0,018	0,24	<0,005	6,3	0,139	0,002	0,118
	105	10,1	<0,01	0,018	<0,02	0,023	0,092	<0,0001	50,8	<0,0005	0,002	<0,0005	0,028	0,019	82,4	0,0002	16,7	0,0010	<0,02	11,6	0,018	0,21	<0,005	9,0	0,136	0,002	0,035
	66	40,6	0,04	0,018	<0,02	<0,02	0,116	<0,0001	70,2	<0,0005	<0,001	0,0009	0,028	0,022	87,6	<0,0001	18,2	0,0020	<0,02	13,3	0,007	0,24	<0,005	14,0	0,176	0,002	0,021
38	97	17,3	<0,02	0,034	<0,02	0,024	0,079	<0,0001	55,5	<0,0005	0,001	0,0006	0,023	0,021	72,6	0,0001	20,0	0,0019	<0,02	9,64	0,013	0,20	<0,005	5,2	0,143	0,003	0,015
	93	13,5	<0,02	0,036	<0,02	0,025	0,080	<0,0001	58,1	<0,0005	<0,001	<0,0005	0,025	0,021	74,4	0,0001	19,6	0,0017	<0,02	10,23	0,010	0,18	<0,005	7,3	0,145	0,003	0,013
	86	33,3	<0,02	0,040	<0,02	0,025	0,111	<0,0001	72,7	<0,0005	<0,001	<0,0005	0,023	0,021	78,1	<0,0001	21,8	0,0015	<0,02	11,70	0,008	0,22	<0,005	11,4	0,176	0,003	0,017
41	93	27,5	0,15	0,018	<0,02	0,026	0,089	<0,0001	76,0	<0,0005	<0,001	<0,0005	0,021	0,017	78,8	<0,0001	23,9	0,0037	<0,02	10,0	0,006	0,18	<0,005	5,0	0,165	0,002	0,005
	98	28,7	0,20	0,024	<0,02	0,031	0,090	<0,0001	70,6	<0,0005	<0,001	<0,0005	0,023	0,016	77,2	<0,0001	22,5	0,0029	<0,02	10,1	0,006	0,18	<0,005	6,3	0,157	0,003	0,014
	94	36,0	0,08	0,029	<0,02	0,050	0,131	<0,0001	70,0	<0,0005	<0,001	<0,0005	0,023	0,019	76,3	<0,0001	23,2	0,0017	<0,02	11,17	0,009	0,16	<0,005	10,3	0,177	0,002	0,030
45	96	32,7	0,29	0,038	<0,02	0,038	0,207	<0,0001	70,5	<0,0005	<0,001	<0,0005	0,019	0,018	74,8	<0,0001	22,8	0,0178	<0,02	10,30	0,014	0,18	<0,005	4,8	0,172	0,002	0,023

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	102	38,9	0,24	0,027	<0,02	<0,02	0,133	<0,0001	79,1	<0,0005	0,002	<0,0005	0,021	0,019	79,8	<0,0001	23,4	0,0092	<0,02	11,11	0,010	0,18	<0,005	6,2	0,184	0,003	0,008
	104	44,4	0,19	0,035	<0,02	0,039	0,132	<0,0001	77,4	<0,0005	<0,001	<0,0005	0,021	0,020	77,9	<0,0001	24,3	0,0024	<0,02	12,32	0,012	0,19	<0,005	9,4	0,188	0,002	0,017
49	84	23,8	0,09	0,015	<0,02	<0,02	0,124	<0,0001	93,8	<0,0005	<0,001	<0,0005	0,023	0,021	98,7	<0,0001	26,5	0,0300	<0,02	12,5	0,005	0,20	<0,005	6,9	0,215	0,003	0,008
	96	23,3	0,83	0,027	<0,02	0,023	0,102	<0,0001	79,7	<0,0005	<0,001	<0,0005	0,021	0,027	87,8	0,0002	21,8	0,0010	<0,02	10,5	0,007	0,24	<0,005	5,4	0,204	0,003	0,006
	85	50,2	<0,1	0,031	<0,02	0,032	0,132	<0,0001	83,9	<0,0005	<0,001	0,0006	0,022	0,026	91,5	<0,0001	23,9	0,0049	<0,02	13,1	0,004	0,22	0,007	9,7	0,206	0,002	0,019

* … not determined

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Table 13. mg/l in the eluates from columns of sludge-treated soil after addition of Fe-treated manure

Days	ml	NO_3-N	NH_4-N	**Al**	**As**	**B**	**Ba**	**Be**	**Ca**	**Cd**	**Co**	**Cr**	**Cu**	**Fe**	**K**	**Li**	**Mg**	**Mn**	**Mo**	**Na**	**Ni**	**P**	**Pb**	**S**	**Sr**	**V**	**Zn**
11	74	42,1	0,08	0,035	<0,02	<0,02	0,144	<0,0001	86,4	<0,0005	<0,001	<0,0005	0,027	0,021	106,5	<0,0001	25,8	0,0017	<0,02	16,8	0,008	0,21	0,005	12,3	0,178	0,003	0,002
	98	39,0	0,17	0,026	<0,02	0,023	0,135	<0,0001	96,8	<0,0005	0,001	0,0006	0,035	0,023	100,3	<0,0001	25,8	0,0023	<0,02	15,5	0,010	0,24	<0,005	13,1	0,240	0,003	0,013
	86	31,4	0,04	0,036	<0,02	<0,02	0,098	<0,0001	67,5	<0,0005	<0,001	0,0007	0,025	0,030	80,4	<0,0001	19,7	0,0014	<0,02	12,1	0,007	0,26	<0,005	10,6	0,171	0,002	0,006
18	45	34,6	0,06	0,024	<0,02	0,022	0,091	<0,0001	59,6	<0,0005	<0,001	<0,0005	0,026	0,022	103,2	0,0001	26,3	0,0033	<0,02	16,4	0,010	0,14	<0,005	19,0	0,181	0,002	0,009
	50	35,8	0,05	0,022	<0,02	0,021	0,106	<0,0001	62,4	<0,0005	<0,001	<0,0005	0,024	0,028	96,7	<0,0001	24,5	0,0022	<0,02	15,0	0,006	0,18	<0,005	13,4	0,182	0,003	0,009
	50	28,2	0,06	0,037	<0,02	<0,02	0,093	<0,0001	47,0	0,0005	0,001	<0,0005	0,027	0,024	86,5	<0,0001	18,4	0,0015	<0,02	13,1	0,007	0,20	0,008	8,4	0,113	0,004	0,003
22	63	31,2	0,07	0,019	<0,02	0,022	0,088	<0,0001	62,0	<0,0005	<0,001	0,0005	0,027	0,023	94,8	0,0002	23,0	0,0020	<0,02	15,1	0,007	0,15	<0,005	16,8	0,168	0,002	0,006
	94	34,3	0,08	0,027	<0,02	0,024	0,103	<0,0001	79,9	<0,0005	<0,001	<0,0005	0,026	0,020	95,8	<0,0001	25,1	0,0015	<0,02	14,0	0,009	0,21	0,005	12,9	0,200	0,002	0,010
	64	33,4	0,35	0,024	<0,02	0,020	0,097	<0,0001	62,4	<0,0005	<0,001	0,0006	0,027	0,025	85,8	0,0002	20,6	0,0013	<0,02	13,6	0,009	0,22	<0,005	13,5	0,155	0,003	0,009
24	54	24,0	0,12	0,038	<0,02	0,061	0,116	<0,0001	71,6	<0,0005	0,001	0,0006	0,030	0,029	96,9	0,0002	22,3	0,0020	<0,02	15,0	0,008	0,20	0,007	19,0	0,194	0,003	0,008
	77	34,4	0,12	0,027	<0,02	0,047	0,122	<0,0001	59,0	<0,0005	<0,001	<0,0005	0,025	0,024	89,6	<0,0001	23,0	0,0017	<0,02	12,5	0,008	0,21	0,005	12,4	0,192	0,003	0,039
	58	22,9	0,05	0,043	<0,02	0,031	0,099	<0,0001	65,3	<0,0005	<0,001	0,0007	0,037	0,028	76,9	<0,0001	19,2	0,0016	<0,02	11,9	0,009	0,25	<0,005	11,5	0,157	0,003	0,028
27	79	28,8	0,08	0,029	<0,02	<0,02	0,097	<0,0001	55,1	<0,0005	0,001	<0,0005	0,028	0,019	96,3	<0,0001	21,7	0,0013	<0,02	14,6	0,005	0,20	<0,005	10,8	0,123	0,003	<0,002
	100	27,5	0,09	0,023	<0,02	0,031	0,104	<0,0001	67,8	<0,0005	<0,001	<0,0005	0,043	0,018	88,5	<0,0001	24,3	0,0012	<0,02	13,0	0,012	0,18	0,005	12,2	0,176	0,003	0,027
	88	27,3	0,15	0,019	<0,02	0,039	0,098	<0,0001	68,3	<0,0005	<0,001	<0,0005	0,023	0,022	73,3	<0,0001	19,8	0,0012	<0,02	10,4	0,010	0,22	<0,005	9,6	0,163	0,003	0,032
35	7	15,9	*	0,047	<0,02	0,054	0,224	<0,0001	86,0	<0,0005	<0,001	0,0008	0,035	0,022	109,0	0,0004	23,6	0,0023	<0,02	27,9	0,059	0,23	<0,005	32,4	0,234	0,003	0,058
	77	29,5	0,02	0,033	<0,02	0,027	0,107	<0,0001	70,5	<0,0005	<0,001	0,0007	0,027	0,023	92,7	<0,0001	20,1	0,0014	<0,02	13,2	0,015	0,24	<0,005	12,4	0,183	0,003	0,020
	23	37,5	0,64	0,031	<0,02	<0,02	0,108	<0,0001	68,6	<0,0005	0,001	0,0005	0,028	0,022	87,8	0,0003	19,4	0,0017	<0,02	13,5	0,012	0,26	<0,005	15,0	0,175	0,004	0,023
38	84	33,8	<0,02	0,047	<0,02	0,035	0,107	<0,0001	73,2	<0,0005	0,001	<0,0005	0,026	0,022	75,4	<0,0001	21,1	0,0012	<0,02	11,63	0,007	0,21	0,007	12,4	0,173	0,004	0,020
	78	29,4	0,06	0,028	<0,02	0,031	0,094	<0,0001	64,3	<0,0005	0,001	<0,0005	0,024	0,020	83,4	<0,0001	23,7	0,0012	<0,02	12,46	0,010	0,17	<0,005	9,8	0,164	0,002	0,009
	72	31,6	<0,02	0,046	<0,02	0,037	0,093	<0,0001	69,7	<0,0005	<0,001	0,0005	0,023	0,023	69,4	0,0002	20,5	0,0012	<0,02	10,15	0,006	0,23	<0,005	9,8	0,166	0,002	0,025
41	90	33,9	0,05	0,021	<0,02	0,049	0,080	<0,0001	65,8	<0,0005	0,002	<0,0005	0,025	0,016	78,4	<0,0001	22,9	0,0014	<0,02	11,5	0,006	0,14	0,005	11,3	0,158	0,003	0,008
	80	34,6	0,05	0,014	<0,02	<0,02	0,121	<0,0001	78,7	<0,0005	0,002	0,0008	0,026	0,021	94,1	<0,0001	22,6	0,0013	<0,02	13,3	0,010	0,22	<0,005	10,4	0,201	0,002	0,012
	57	41,1	0,04	0,026	<0,02	0,028	0,099	<0,0001	68,1	<0,0005	<0,001	<0,0005	0,022	0,022	72,2	<0,0001	20,6	0,0012	<0,02	11,11	0,009	0,19	<0,005	9,7	0,161	0,002	0,011
45	67	39,3	0,02	0,019	<0,02	0,043	0,082	<0,0001	67,1	<0,0005	<0,001	0,0007	0,024	0,015	76,3	<0,0001	21,8	0,0014	<0,02	11,3	0,006	0,15	<0,005	10,1	0,154	0,002	0,014

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	89	11,6	<0,15	0,072	<0,02	0,023	0,100	<0,0001	73,4	<0,0005	0,001	0,0012	0,036	0,057	103,2	0,0002	21,0	0,0063	<0,02	15,1	0,009	0,30	0,007	12,0	0,173	0,004	0,010
	78	40,4	0,21	0,024	<0,02	0,022	0,092	<0,0001	73,8	<0,0005	<0,001	<0,0005	0,021	0,016	79,0	<0,0001	25,2	0,0192	<0,02	10,4	0,002	0,15	<0,005	7,3	0,175	0,003	0,007
49	69	23,6	<0,1	0,022	<0,02	<0,02	0,102	<0,0001	74,0	<0,0005	<0,001	<0,0005	0,025	0,021	93,8	<0,0001	24,7	0,0022	<0,02	13,6	0,005	0,18	<0,005	10,7	0,180	0,002	0,013
	74	24,9	<0,1	0,022	<0,02	<0,02	0,107	<0,0001	80,0	<0,0005	<0,001	0,0006	0,021	0,020	91,3	<0,0001	22,9	0,0061	<0,02	12,2	0,003	0,19	<0,005	7,9	0,192	0,002	0,015
	15	18,1	<0,3	0,286	<0,02	<0,02	0,127	<0,0001	86,8	<0,0005	<0,001	0,0013	0,030	0,220	89,9	0,0003	21,8	0,0091	<0,02	12,9	0,007	0,45	0,007	11,2	0,224	0,003	0,023

* … not determined